

DYNAMIC MODELLING AND OPTIMAL CONTROL OF A MULTICOMPONENT BATCH PACKED DISTILLATION COLUMN

Hatice Ceylan*, Canan Özgen**

Chemical Engineering Department, Middle East Technical University, Ankara 0631, Turkey

(* e119196@metu.edu.tr, ** cozgen@metu.edu.tr)

A dynamic model for the simulation of the batch operation of a multicomponent packed distillation column is developed. The accuracy of simulation code is verified by using two types of mixtures (cyclohexane-n-heptane-toluene and ethanol-water) theoretically and experimentally. An optimum internal reflux ratio profile is searched to maximize the distillate amount of a specified concentration for a given time by using capacity factor approach using MATLAB software.

Keywords: Process modelling, batch and semi-batch process control, nonlinear process control, modelling for control optimization

1. INTRODUCTION

Due to high efficiency, high capacity to resist to corrosive materials, low pressure drop and small liquid hold-up packed columns are preferred compared to plate columns in distillation (Perry et al., 1997). The choice of the type of operation of distillation, either batch or continuous, depends on the feed amount and on the characteristics of the feed components. Nowadays, batch distillation is more commonly used due to its convenience for low volume fine chemicals and biochemicals (Betlem, 2000) and due its advantages like "flexibility, high product purity and possibility of multiple fraction operation" (Li et al., 1998).

Packed distillation column design can be achieved by considering packing either as a continuous media or as transfer units. Gorak and Vogelpohl (1985) have shown that use of Height Equivalent to a Theoretical Plate (HETP) causes very poor results. Krishnamurthy and Taylor (1985) considered packing as a continuous media. This method had been used by several authors like Karlström et al. (1992), Pathwardhan and Edgar (1993), Mori et al. (1999), and Repke et al. (2004). It is very difficult to determine the composition changes with time throughout the batch distillation columns at optimum reflux ratios by performing experiments or by taking data from the column due to complex column dynamics (Kreul et al., 1999). As Fieg et al. (1994) stated, the dynamic behavior of an actual distillation column can be predicted at different operating conditions by using the simulation algorithm.

In this study, a dynamic model for the simulation of a batch packed distillation column with random packing material for multicomponent mixtures at low operating pressures is developed. The optimum reflux ratio profile of the multicomponent batch packed distillation column system is

determined by using capacity factor (CAP) approach developed by Luyben (1988) in order to maximize the amount of distillate of a specified concentration for a given time.

2. DYNAMIC MODELING

In the proposed batch packed distillation column model, following assumptions are considered (Yıldız (2002), Ceylan (2007)); negligible vapor hold-up throughout the system, equimolar counter diffusion between the phases, adiabatic column operation and negligible temperature change throughout the packed section, dominance of overall vapor phase mass transfer coefficient to individual liquid and vapor film mass transfer coefficients, negligible effect of maldistribution of liquid and vapor flow rates, constant liquid hold-up in the reflux drum, change of composition only in z direction, and no radial variation, perfect mixing of compositions throughout the system and total condenser.

A schematic view of the studied batch packed distillation column with its reboiler, condenser, reflux drum and tanks is shown in Figure 1. The parameter k in Figure 1 defines the differential element starting with k = 1 at reboiler. P₁ and P₂ are the product tanks and S₁ indicates the slop cut tank. In the present study, discrete element concept is used instead of HETP for dynamic modelling where the packed section is divided by NT into segments of Δz height (Δz = (Height of packed section) / NT) whose value can be changed arbitrarily and will have an effect on modeling accuracy.

Model equations are given below as;

Conservation equations; for j = 1,...,NC liquid and vapor phase component mass balances are;

$$M'_{L,j} \frac{\partial x_j}{\partial t} = L \frac{\partial x_j}{\partial z} - K_{Vj} A a_e (y_j^* - y_j) \quad (1)$$

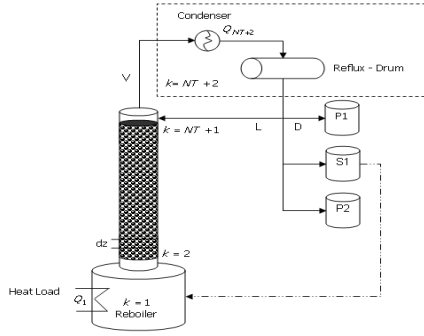


Fig. 1. Schematic view of studied batch packed distillation column.

$$0 = -V \frac{\partial y_j}{\partial z} + K_{Vj} A a_e (y_j^* - y_j) \quad (2)$$

with initial and boundary conditions:

$$x(z,0) = x(z); y(z,0) = y(z); y(0,t) = y_B \quad (3)$$

The approach used by Attarakih et al. (2001) is applied to convert partial differential equations (PDEs) to ordinary differential equations (ODEs) as follows:

For $i = 1, \dots, DL$

$$\frac{\partial x}{\partial z} = \frac{x_{i+1} - x_i}{\Delta z}; \frac{\partial y}{\partial z} = \frac{y_{i+1} - y_i}{\Delta z} \quad (4)$$

$$y^* = \frac{y^*(x_i) + y^*(x_{i+1})}{2}, y = \frac{y_{i-1} + y_i}{2} \quad (5)$$

Then, Equations 1 and 2 can be written as:

$$\frac{dx_{i,j}}{dt} = \frac{L}{M'_L} \frac{x_{i+1,j} - x_{i,j}}{\Delta z} + \frac{K_{Vj} A a_e}{2M'_L} \left((y_{i-1,j} + y_{i,j}) - (y^*(x_{i,j}) + y^*(x_{i+1,j})) \right) \quad (6)$$

$$y_{i+1,j} = - \frac{K_{Vj} A a_e \Delta z}{V} \left(\frac{y_{i-1,j} + y_{i,j}}{2} - \frac{y^*(x_{i,j}) + y^*(x_{i+1,j})}{2} \right) + y_{i,j} \quad (7)$$

Liquid phase component mass balance at the reboiler ($k = 1$) and at the reflux – drum – condenser ($k = NT + 2$):

$$\frac{dx_{1,j}}{dt} = [L(x_{2,j} - x_{1,j}) - V(y_{1,j} - x_{1,j})] / M_1 \quad (8)$$

$$\frac{dx_{NT+2,j}}{dt} = [V(y_{NT+2,j} - x_{NT+2,j})] / M_{NT+2} \quad (9)$$

With external reflux ratio ($R = L/D$), flow rate of distillate at finite external reflux ratio is:

$$D = \frac{Q_1 - \sum_{n=1}^{NT+2} \frac{d(M_n h_n)}{dt}}{(R+1)H_{NT+1} - Rh_{NT+2}} \quad (10)$$

Liquid flow rate at total reflux ($D = 0$) is:

$$L = \frac{Q_1 - \frac{d(M_1 h_1)}{dt} - \frac{d(M_{NT+2} h_{NT+2})}{dt}}{(H_{NT+1} - h_{NT+2})} \quad (11)$$

Onda correlations (Wang et al., 2005) are:

$$k_V = c \left(\frac{D_V}{a_p d_p^2} \right) \text{Re}_V^{0.7} \text{Sc}_V^{1/3} \quad (12)$$

$$\frac{a_e}{a_p} = 1 - \exp \left[-1.45 \left(\frac{\sigma_c}{\sigma_L} \right)^{0.75} \text{Re}_L^{0.1} \text{Fr}_L^{-0.05} \text{We}_L^{0.2} \right] \quad (13)$$

Liquid molar hold-up correlations below loading point and above loading point are given by (Stichmair, et al., 1989):

$$h_o = 0.555 \text{Fr}_L^{1/3} \quad (14)$$

$$h_L = h_o \left[1 + 20(\Delta p_{irr} / (z \rho_L g))^2 \right] \quad (15)$$

Linear pressure drop profile can be written as:

$$P(z) = P_B + \Delta z * \frac{\Delta P_{irr}}{Z} \quad (16)$$

In the modeling, dry, irrigated and flooding pressure drop correlations (Stichmair et al., 1989) are used whereas equal fugacities are considered for liquid and vapor phases. For hydrocarbons at low pressures, to predict VLE relationships Peng Robinson Equation of State (PR – EOS) is utilized for both, vapor and liquid phases. On the other hand, PR – EOS must not be used to predict vapor liquid equilibrium for liquid phase of polar mixtures (Sandler, 1999). Bahar (2007) showed that, the use of Non Random-Two Liquid (NRTL) activity coefficient model gives best results for the prediction of compositions of polar mixtures. Therefore, for polar mixtures NRTL activity coefficient will be used to predict liquid phase and, PR – EOS will be used to predict vapor phase compositions at equilibrium. Moreover, in this study, prediction of physical parameters like viscosity and surface tension are done at low pressures by most commonly used correlations for hydrocarbons, and for polar mixtures, these parameters are kept constant. Model equations are solved using MATLAB software by using Euler's method to integrate state equations (Yıldız, 2002).

3. OPTIMAL CONTROL

Optimization of reflux ratio profile to maximize distilled product amount in a multicomponent batch packed distillation column is performed by maximizing CAP which was developed by William L. Luyben in 1971 and defined as “the total specification products produced (P_i) divided by the total time of batch” (Luyben, (1988), Ceylan and Özgen (2007)). Formulation of the problem necessitates the inputs as; column, feed mixture and product specifications, heat load and initial values (reflux ratio, pressure in the reboiler, dummy variables etc.) by using the following representation (Bahar, 2007):

$$CAP = f(x) = \frac{\sum_{i=1}^{NC} M_{P_i}}{t_T + 0.5} \quad (17)$$

$$\begin{aligned} \max f(x) \text{ subject to:} \\ 0 \leq R_p \leq 1 \end{aligned} \quad (18)$$

where R_p is the internal reflux ratio (L/V).

As Luyben (1988) and Bonny (1999) stated recycling of holdups of the slop cut tanks do not have any significant effect on CAP in plate distillation columns. However, it increased the product amount by 16% for multicomponent batch plate distillation columns. In this study, also the recycling of molar hold-up of slop cut tanks for the next batch is examined to increase the product amount and to analyze the effect of recycle on CAP in packed distillation columns.

4. CASE COLUMN AND EXPERIMENTAL SET UP

Simulation algorithm is checked qualitatively as in the case of Hitch and Rousseau (1988) by comparing nine simulation runs with varying one parameter at each simulation run. A case column is considered as having the same specifications of the column and feed used in the study of Mujtaba and Macchietto (1993). The column specifications in the study of Mujtaba and Macchietto (1993) are given as tray numbers, boil up rate, hold-ups of the trays, hold-ups of condenser-reflux-drum and pressure drop throughout the column. Since the column used in the study of Mujtaba and Machietto (1993) is a plate distillation column, some more specifications has to be included as inputs to the simulation program for the packed distillation column. Thus, irrigated and flooding pressure drops are evaluated by the simulation code using the top pressure given in the study of Mujtaba and Machietto (1993). Also, the design gas rate is selected to be 38% of the flooding rate for a column of 0.35m in diameter with packed height of 1.5m (Table 1).

Table 1 Atmospheric column, feed and used packing material specifications

Maximum Boil up Rate (kmol/h)	2.75
Fresh Feed Amount (mol)	2930
Initial mole fractions at reboiler	
Cyclohexane	0.407
n- Heptane	0.394
Toluene	0.199
Packing material	Ceramic ring Raschig
Diameter(m)	0.010
Specific surface area of the packing (m^{-1})	472
Porosity	0.655

Simulation algorithm is verified quantitatively, by carrying out experiments on a lab scale packed atmospheric distillation column by using 60% EtOH and 40% H_2O mixture [mol/mol]. Ethanol of $\geq 99.99\%$ (w/w) purity (supplied by Merck) and pure water are used to prepare the mixture. The experimental set – up used in this study consists

of a packed distillation column made of glass and with an ID of 5cm and with 0.4m packing height. Column is filled with 5 mm diameter glass Raschig Rings. 1000 ml solution is used as the polar mixture. The mixture is initially 23 moles. Heat load is chosen as 500 W (1800000 J/h). Flow rate of condenser is 1.4 l/min. In order to reduce heat loss to the environment, reboiler and packed column are insulated.

5. RESULTS AND DISCUSSION

The selection of the size of the incremental packing bed height of the batch packed distillation column, Δz is important for the accuracy of the results. Thus, first the effect of NT (15, 35, 40 and 50) on the accuracy of the results is investigated by using a case study found from the literature (Yıldız (2002)) and found that NT = 40 is adequate for the accuracy of the results for hydrocarbon mixtures of cyclohexane, n-heptane and toluene.

5.1 Verification of Simulation Code for Hydrocarbon Mixtures

In batch distillation operation, there are two important periods: start-up period and production period. In the production period the composition in the condenser changes since distillate is withdrawn from the column. The written simulation program can handle both – start-up and production periods and solves the state and algebraic equations to find the vapor and liquid compositions, change of temperatures, hold-ups, liquid and flow rates during start-up and production periods. In the simulation runs, the column is first operated at total reflux at the end of which steady state is reached and then, production period is started.

Similar to the study done by Hitch and Rousseau (1988) internal reflux ratio (0.7, 0.75 and 0.8), height of packing (1.5m, 2m and 3m), condenser hold-up (20moles, 100 moles and 200 moles) and heat load ($5 \cdot 10^7$, $6.5 \cdot 10^7$ and $8.15 \cdot 10^7$ J/h) given to the system are changed and the effect of these variables on distillate compositions are analyzed and checked whether expected behaviors are achieved or not. Base run is done using reflux ratio as 0.8, height of packing 1.5m, condenser holdup as 20moles and heat load as $8.15 \cdot 10^7$ J/h).

Figure 2 demonstrates the effect of internal reflux ratio on distillate compositions. As seen the highest purity of cyclohexane is obtained after one hour of total reflux period. Furthermore, in terms of n-heptane, it is observed that, maximum concentrations are decreasing as reflux ratio decreases. Operation times decrease as internal reflux ratio decreases. This trend is an expected one in a batch distillation system as found also by Hitch and Rousseau (1988).

As height of packing is changed from 1.5m to 2m and 3m to observe its effect on separation, while keeping other parameters constant at base run. The change in distillate compositions is shown in Figure 3. The total effective interfacial area for mass transfer increases as the height of packing increases which increases the total mass transferred. Thus, the maximum concentrations of the lightest (in total reflux period) and second lightest components (in production period) increase as packed height increases as expected.

The effect of condenser hold-ups on the distillate compositions are given in Figure 4. The time necessary to reach a steady state value increases as the amount of condenser hold-up increases in start period. Steady state compositions are nearly same for 20, 100 and 200 moles condenser hold-ups. In product period, the separation efficiency of column decreases as the amount of condenser hold-up increases.

The effect of heat loads is given in Figure 5. The highest purity of cyclohexane is obtained at start-up period. The maximum purities of the n-heptane do not change very much as the heat load is changed during the production period. However, the operation time is affected by the heat load given to the reboiler. As the heat load decreases, operation time for separation increases as expected.

Considering the effect of four parameters on the operations of batch packed distillation system, the expected effects are observed which verifies qualitatively simulation code prepared.

5.2 Experimental Verification of Simulation Code for Polar Mixtures

Verification of simulation code for polar mixtures is achieved quantitatively by comparing the results of simulation with the experimental findings. The experiments are performed for a binary mixture of ethanol – water with total reflux ratio for 5 hrs and then with an external reflux ratio of 0.5 for 1 hr. Reid et al. (1987) stated that, the most accurate results are predicted by using Wagner equation. Therefore, experimental findings are compared with the simulation results using Wagner Equations (Reid et al. (1987)) for the vapor pressure of the components in terms of temperature.

The comparison of the experimentally obtained dynamic ethanol concentrations of the distillate and their calculated values with simulation are given in Figure 6. As expected, the highest purity of liquid ethanol composition is obtained at total reflux both theoretically and experimentally. Thus, experimental verification of the simulation code using polar mixture has been successfully done considering the behavior of the distillate composition with time with an error of 12% in experimental composition values considering Wagner equation in the simulation for pressure dependence. The error found may be due to the constant assumption of NRTL parameters and physical properties at 20°C.

5.3 Determination of Optimum Reflux Ratio and Effect of Slop Recycling on CAP and Separation Efficiency

During the production period of batch distillation, distilled material is collected in separate storage tanks due to changes in with time. The lightest product (mostly cyclohexane) and the second lightest product (mostly n- heptane) are collected into P₁ and P₂ tanks, respectively. In between, the “off-spec” material as waste is collected in S₁. The heaviest product, which includes toluene mostly, is withdrawn from the reboiler after distillation stops. Thus, at the final stage, the hold-ups of the packed section and condenser are also drained and collected in the reboiler, P₃. Using Equation 17 and 18

and using optimization toolbox in MATLAB software, optimum internal reflux ratio is calculated with time interval and shown in Figure 7 (Ceylan and Özgen (2007)).

The desired purities for cyclohexane and n – heptane are 0.9 and 0.8, respectively. The operation is continued for an internal reflux ratio of 0.889 for about 2.11 hours to collect the distillate in the first product tank, P₁, until distillate composition drops below 0.9 with a CAP value of 175.71 mol/hr. Then, the product, which is “off-spec”, is collected in slop-cut tank S₁ for a certain period of time 2.77 hours with

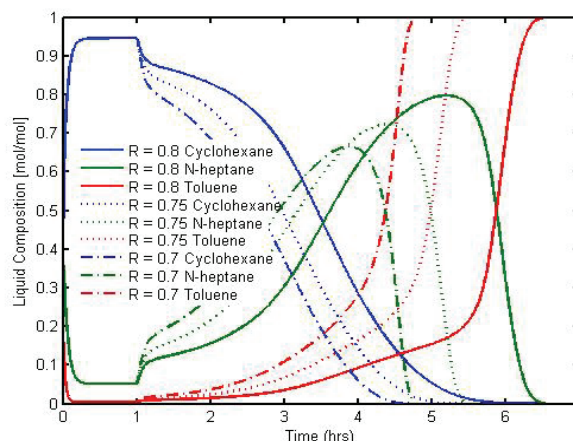


Fig. 2. Effect of internal reflux ratio on the distillate compositions.

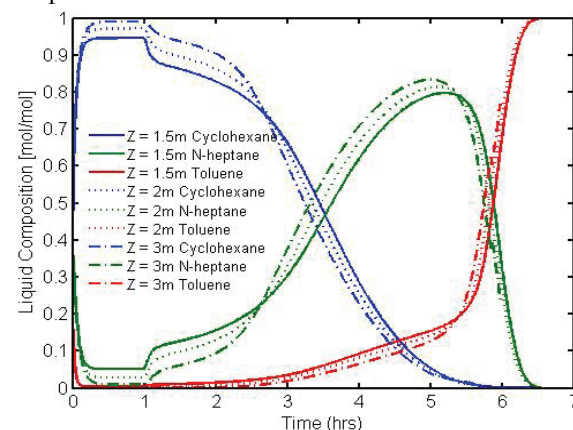


Fig. 3. Effect of packing height on the distillate compositions.

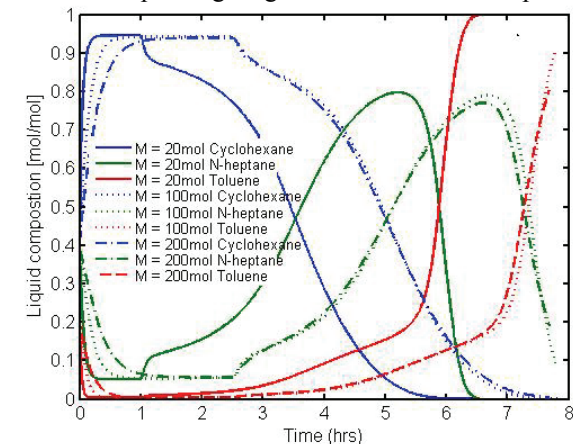


Fig. 4. Effect of condenser hold-ups on the distillate compositions.

an internal reflux ratio of 0.825 until the second lightest component composition reaches 0.8. Then, the operation continues with an internal reflux ratio of 0.794 until the toluene composition in the reboiler reaches 0.69 for about 0.72 hours with a CAP value of 235.07 mol/hr. The third compound, toluene is collected in the reboiler. According to simulation results, the purity of the heaviest compound, toluene is 0.69 at reboiler after 6.6 hours operation. However, the purity of toluene decreases to 0.66374 due to the addition of the hold-ups of the packed section and condenser at the end of the operation.

Initially, the amount of the mixture fed to the reboiler for separation is 2930 moles. After optimum operation by using reflux ratio profile, 56.96% of the mixture (1669 moles) is separated into the components with desired purities in the first phase of the distillation operation. The molar hold-up of slop cut tank, S_1 , is 1261 moles and is large enough not to be wasted. One of the ways to maximize distilled product amount is to recycle hold-ups of slop cut tank for the next batch distillation operation. Therefore, the contents of the slop cut tank are fed to the reboiler for the two proceeding batches. The summary of the results of recycling processes are shown in Table 2.

As a result of the study, CAP for the optimization with recycling is calculated as 188.86 mol/h. In the calculation of CAP, time necessary for emptying and charging the feed to the reboiler is included for two times (0.5×2 hrs) because in the 3rd operation, reboiler contents are distilled. The total operation time of separation process for fresh feed and further distillations are included. It is observed that CAP which is 235.07 mol/hr without recycling process decreases to 188.86 mol/hr. This is because; the time necessary for separation of fresh feed and slop cut tanks increases beside the molar hold-ups of the product tanks. Moreover, the overall separation efficiency is calculated as 72.96% for the fresh feed with recycle whereas it is 56.96% without recycle. Thus, there is a 28% increase in separation efficiency with recycling. Therefore, it can be concluded that recycling of the hold-ups of the slop cut tanks increases the separation efficiency of the process, significantly.

7. CONCLUSIONS

The dynamic model of a multicomponent batch packed distillation column with random packing material is developed and checked by a simulation code and by experiments. The optimum reflux ratio profile is found to maximize the amount of products. It is found that, optimal reflux ratio profile is a piecewise constant function of time. In recycling of hold-ups of slop cuts to collect the lightest compound more effectively for the proceeding batch operation it is found that, one recycling is enough with an increase efficiency of separation about 28%. Further recycle process does not give any further improvements on maximizing the amount of distillate with the specified desired purity of compounds.

8. ACKNOWLEDGMENT

Financial support of The Scientific and Technological Research Council of Turkey (TUBITAK) is acknowledged.

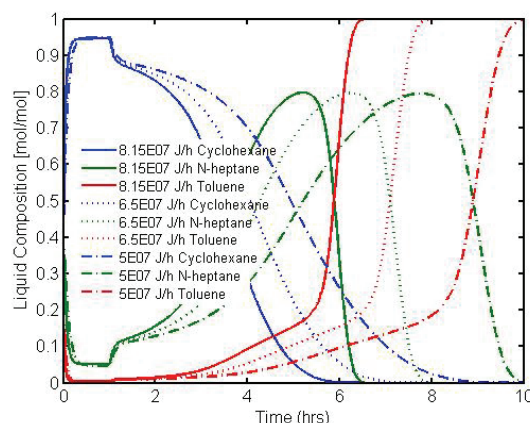


Fig. 5. Effect of heat loads on the distillate compositions.

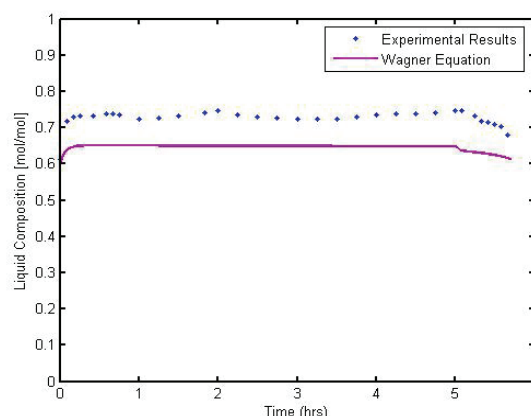


Fig. 6. Comparison of results obtained by simulation and experiments.

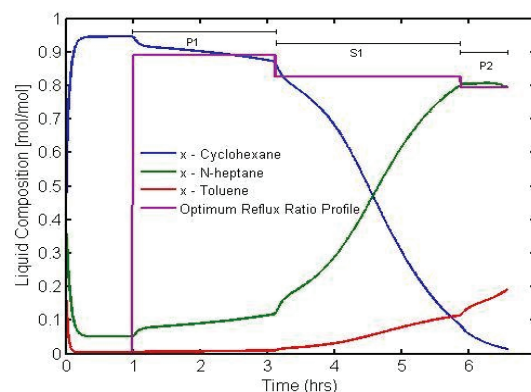


Fig. 7. The liquid distillate compositions obtained under optimum reflux ratio profile.

Table 2 Summary of the operations with recycling

Tank	Compound	Amount of Hold-ups [mol]			
		Operation			Total
		1 st	2 nd	3 rd	
P ₁	Cyclohexane	634.3	456.9	12.0	1103.2
P ₂	N- heptane	369.9	0.0	0.0	369.9
P ₃	Toluene	664.8	0.0	0.0	664.8
S ₁	“Off – spec” material	1261.0	804.1	792.1	792.1
Total					2930.0

NOMENCLATURE

A	Tower cross-sectional area [m ²]
a _c , a _p	Effective interfacial and specific packing surface areas per unit volume of packing volume [1/m]
c	Packing specific constant
D	Distillate flow rate [mol/hr]
D	Diffusion coefficient [m ² /s] in Equation 12
d _p	Particle diameter [m]
h, H	Liquid and vapor mixture enthalpies [J/mol]
h _o	Liquid holdup below the loading point [m ³ /m ³]
h _L	Liquid holdup above the loading point [m ³ /m ³]
k	Mass transfer coefficient [m/s]
K	Overall mass transfer coefficient [mol/(h.m ² .mol frac)]
L, V	Liquid and vapor flow rates [mol/hr]
M	Molar hold-up [mol]
M'	Molar hold-up / height [mol/m]
P	Pressure [Pa] in Equations 15 and 16
Q	Heat load [J/hr]
t	Time [hr]
T	Temperature [K]
x _z , y	Liquid and vapor fractions [mol/mol]
y	Equilibrium vapor mole fraction [mol/mol]
z	Packing height at any time [m]
Z	Total packing height [m]
Greek Letters:	
ρ	Density [kg.m ⁻³]
σ	Surface tension [mN/m]
Subscripts:	
B	Reboiler
c	Critical
i	Discretization level number index
irr	Irrigated
j	Component number index
k	Stage index
T	Total

REFERENCES

- Attarakih, M., Fara, A. D. and Sayed, S. (2001). Ind. Eng. Chem. Res., *Dynamic modelling of a packed-bed glycerol-water distillation column*, **40**, pp.4857 – 4865.
- Bahar, A. (2007). *Control and simulation studies for a multicomponent reactive batch distillation column*, PhD. Thesis, Middle East Technical University, Ankara.
- Betlem, B.H.L. (2000). Chemical Engineering Science, *Batch Distillation column low – order models for quality program control*, **55**, pp. 3187 – 2194.
- Bonny, L. (1999). Ind. Eng. Chem. Res. *Multicomponent batch distillations: study of operating parameters*, **38**, pp. 4759 – 4768.
- Ceylan, H. (2007). *Control and Simulation Studies for a Multicomponent Batch Packed Distillation Column*, M. Sc. Thesis, Middle East Technical University, Ankara.
- Ceylan H. and Özgen C., 05 – 07 September (2007). TOK'2007 Otomatik Kontrol Ulusal Toplantısı, *Çok bileşenli kesikli dolgulu damıtma kolonunun optimal denetimi*, Sabancı Üniversitesi, İstanbul.
- Converse, A. O. and Gross, G. D. (1963). Ind. Eng. Chem. Res., *Optimal distillate – rate policy in batch distillation*, **2**, No. 3, pp. 217 – 221.
- Coward, I. (1967). Chemical Engineering Science, *The time – optimal problem in binary batch distillation*, **22**, pp.503 – 516.
- Fieg, G., Wozny, G., Jeromin, L. and Kruse, C. (1994). Chem. Eng. Technology, *Comparison of the dynamic behaviour of packed and plate distillation columns*, **17**, pp. 301 – 306.
- Gorak, A., Vogelpoh, A. (1985). Separation Science and Technology, *Experimental study of ternary distillation in a packed column*, **20** (1), pp.33–6.
- Hitch, D.M. and Rousseau, R. W. (1988). Ind. Eng. Chem. Res., *Simulation of continuous – contact separation processes: multicomponent batch distillation*, **27**, pp. 1466 – 1473.
- Karlström, A., Breitholtz, C. and Molander, M. (1992). Chem. Eng. Technology, *Heat, Mass and Momentum Transfer in Packed Bed Distillation Columns*, **15**, pp. 1 – 10.
- Kreul, L. U., Górak, A., Barton, P.I., (1999). AIChE Journal, *Dynamic rate – based model for multicomponent batch distillation*, **45**, No.9, pp. 1953 – 1962.
- Krishnamurthy, R., and Taylor, R. (1985). Ind. Eng. Chem. Process Des. Dev., *Simulation of packed distillation and absorption columns*, **24**, pp. 513 – 524.
- Li, P., Hoo H. P., Wozny G., (1998). Chem. Eng. Technol., *Efficient simulation of batch distillation processes by using orthogonal collocation*, **11**, pp. 853 – 862.
- Luyben, W. L. (1988). Ind. Eng. Chem. Res., *Multicomponent batch distillation I. ternary systems with slop recycle*, **27**, pp. 642 – 647.
- Mori, H., Ito, C., Oda, A. and Aragaki, T. (1999). Journal of Chemical Engineering of Japan, *Total reflux simulation of packed column distillation*, **32**, No. 1, pp. 69 – 75.
- Mujtaba, I.M. and Macchietto, S. (1993). Computers Chem. Engng., *Optimal Operation of Multicomponent Batch Distillation – Multiperiod Formulation and Solution*, **17**, No.12, pp.1191 – 1207.
- Patwardhan, A. A. and Edgar, T. F. (1993). Ind. Eng. Chem. Res., *Nonlinear model predictive control of a packed distillation column*, **32**, pp. 2345 – 2356.
- Perry, R.H., Green, D.W., Maloney, J.D. (1997). *Perry's chemical engineer's handbook*, 7th edition, McGraw-Hill, New York.
- Reid, R.C., Prausnitz, J.M., Poling B.E. (1987). *The Properties of Gases and Liquids*, 4th Edition, McGraw-Hill: New York.
- Repke, J. U., Villain, O., Günter, W. (2004). Computers and Chemical Engineering, *A nonequilibrium model for three-phase distillation in a packed column: modelling and experiments*, **28**, pp. 775 – 780.
- Sandler, S. I. (1999). *Chemical and engineering thermodynamics*, 3rd Edition, John Wiley & Sons, Inc.
- Stichlmair, J., Bravo J.L., Fair, J.R. (1989). Gas Separation and Purification, *General model for the prediction of pressure drop and capacity of counter current gas / liquid packed columns*, **3** (1), pp. 19 – 28.
- Wang, G. Q., Yuan, X.G., Yu, K. T. (2005). Ind. Eng. Chem. Res., *Review of mass transfer correlations for packed columns*, **44**, pp. 8715 – 8729.
- Yıldız, U. (2002). *Multicomponent batch distillation column simulation and state observer design*, M. Sc. Thesis, Middle East Technical University, Ankara.