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Sensitivity Analysis and Rigorous Simulation of the Three-Phase Distillation Process of Ethanol/Ethyl Acetate/Water Mixture Based on Conceptual Design Principles

Shima Sheibani[1,2], Behrooz Mahmoodzadeh Vaziri[1,3]✉

[1] Department of Chemical Engineering, Qu.C., Islamic Azad University, Quchan, Iran. E-mail: sheybani.shima@yahoo.com
[2] Department of Physics and Science, Aston University, Birmingham, UK. E-mail: sheybani.shima@yahoo.com
[3] Corresponding Author, Department of Chemical Engineering, Ma.C., Islamic Azad University, Mashhad, Iran.
E-mail: behrooz.vaziri@iau.ac.ir

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ABSTRACT

The three-phase distillation operation is one of the most advanced distillation methods for the separation of azeotropic mixtures. In this study, the optimization and rigorous design of the three-phase distillation process of the highly non-ideal mixture of ethanol/ethyl acetate/water was carried out based on conceptual design principles. Initially, the possibility of separating this highly complex mixture (at different concentrations) with four azeotropic points and three distillation regions was evaluated using different purification patterns. According to the results, it was determined that the second distillation region (the region where the stable component is ethanol) is the best one for the separation. Then, the effect of operating pressure and reflux ratio on the conceptual design of the three-phase column was investigated in the optimal distillation region. In the following, based on the conceptual design results, the rigorous simulation of the process was performed. Subsequently, the operational and design variables were optimized using the distinct and simultaneous sensitivity analysis methods. Based on optimized values, ethanol with a purity of 98.7mol% was separated with a duty of 4754.14kW. Finally, with the rigorous design of the column, it was revealed that to reach the mentioned purity, the packed columns with Mellapak 64x packing are smaller in size than the tray columns with sieve trays.

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1. Introduction

Ethyl acetate is a colorless liquid with a slightly sweet odor. This substance is primarily used as a solvent and diluent due to its advantages, including cost efficiency, accessibility, low toxicity, and a pleasant aroma. It is used in the production of printing inks, adhesives, polishes, cleansers, fragrance products, candies, solvents in resins, artificial leather, varnish, ink, photographic films and as a gelling agent in the production of gunpowder. Ethyl acetate is also used in the extraction of oils, antibiotics, resins, gums, and the extraction of caffeine from coffee and tea [1-7].

From the reaction of ethanol with acetic acid, ethyl acetate (the main product) and water (byproduct) are produced. To use the whole acetic acid, ethanol enters the reactor in an extra amount. As a result, the reactor output will have a significant amount of ethanol in addition to the reaction products (ethyl acetate and water), which should be separated for reuse [1,3,6].

Ethanol extraction is a critical step in the process of ethyl acetate purification. Most of the research conducted on the deethanolization of the ethyl acetate process are based on reactive distillation [8-12], pervaporation [13,14], pressure-swing distillation [15,16], extractive distillation [16,17], and hybrid separation processes [16,18].

Reactive distillation is a relatively complex process. This complexity results from a strong interaction between chemical reactions with mass and heat transfer operations and high sensitivity to continuous variables of the column such as reflux ratio [8,9,19]. On the other hand, the use of reactive distillation is partially limited by restrictions such as the operating conditions required for distillation and chemical reaction, the difficulty of controlling the process, and the availability of suitable residence time [9,19]. Another drawback of reactive distillation is associated with the use of basic or homogeneous acidic catalysts, which leads to serious environmental and economic problems. In addition, these catalysts are expensive and require serious considerations to maintain their long-term thermal stability [8,11]. Also, catalysts are usually contaminated through feed impurities [11].

Pervaporation is involved in the problems of intense competition between the components of the mentioned mixture against the membrane. In addition, the possibility of the copermeation of mixture components into the membrane can seriously affect the separation efficiency [13,20].

Pressure-swing distillation is a method used to separate azeotropic or close-boiling mixtures by exploiting how the azeotropic composition shifts with pressure. Although changing the equilibrium properties of an azeotropic mixture by pressure changes is an attractive possibility, this process is faced with various challenges such as high capital and operating costs, high energy consumption, safety concerns, and scale-up problems [15,21].

Extractive distillation is a separation method used to break azeotropes or separate close-boiling mixtures by adding a high-boiling entrainer (solvent) that alters the relative volatilities of the components. However, it also has several disadvantages, such as high energy consumption, solvent handling and recovery, risk of contamination from solvent impurities, and environmental and safety concerns [16,17].

Hybrid techniques are a combination of the separation processes mentioned above. Pervaporation can be applied as hybrid system with reactive distillation to produce high-purity ethyl acetate [22]. The extractive heterogeneous-azeotropic distillation (EHAD) combines the heterogeneous-azeotropic and extractive distillations for the separation of highly non-ideal liquid mixtures [18,23]. Recently extended hybrid systems including the combination of extractive, heterogeneous-azeotropic, pressure swing distillation and pervaporation techniques were used to separate ethyl acetate/water/ethanol mixture [16,18]. Although these techniques are designed to overcome the limitations of each of the separation methods alone, they are involved in severe systemic complexities and interferences.

As environmental challenges become increasingly serious and fossil fuels are running out, implementing a separation method with low energy consumption and environmental pollution becomes increasingly important. In the present study, the three-phase distillation method is used for removing ethanol from the ethyl acetate process (Fig. 1). The reason for this is the various advantages of this method, including the ease of implementation, design, reliability scale up, more complete separation, efficient handling of complex mixtures, and ease of separating the vapor-liquid phases compared to other processes [24,25]. However, the main advantage of the three-phase distillation process is reducing duty, since it is based on boiling at a minimum temperature. In fact, in this process, the creation of an azeotrope with a minimum boiling point allows distillation at lower temperatures. As a result, this method can be used to save energy consumption and to distillate unstable and heat-sensitive mixtures [9, 13, 26]. Although this method has many advantages, it also deals with some challenges, including higher maintenance requirements, control complexity (needing precise control of temperature, pressure, and flow to maintain distinct phases), and the risk of emulsion formation [20,21].

The three-phase distillation process occurs when at least one component of the liquid mixture is miscible in other components. Therefore, in this process, the vapor phase is in contact and equilibrium with two liquid phases in tray or packed column. For this reason, for the separation of the two liquid phases created in this process, the three-phase distillation column is always accompanied by a decanter (Fig. 1) [10,11,27]. Based on the number of insoluble components, a three-phase distillation operation can be performed in two ways: single decanter (if one of the components is insoluble in one of the other components of the mixture) and two decanters (if one of the components is insoluble in two other components) [11].

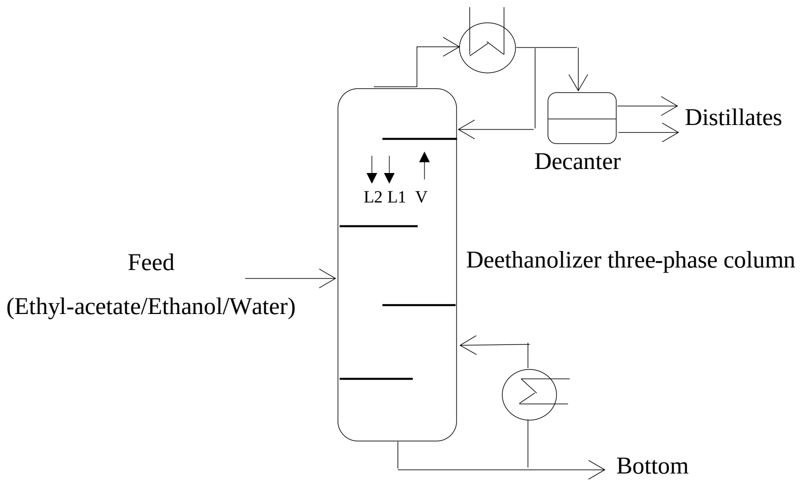

Fig. 1. Proposed three-phase distillation column for the deethanolization of ethyl acetate process

As stated above, the three-phase distillation operation can be an attractive and efficient process for the separation of azeotropic mixtures that are needed today by many chemical and petrochemical industries. On the other hand, distillation columns and their support facilities cover about one-third of investment costs and more than half the energy costs of an operational unit [24,25]. As a result, the main focus of the current paper is the optimization and rigorous design of a three-phase distillation column for deethanolization based on the conceptual design principles to reduce the cost of the ethyl acetate process. According to our knowledge, the proposed method is used for the first time for removing ethanol from the ethyl acetate unit and has not been reported in published papers.

2. Conceptual design

The design and optimization of three-phase distillation towers is a challenging issue due to heterogeneity in the liquid phase. In this regard, the larger the number of azeotropic points, the more difficult the three-phase separation operation becomes due to the creation of limiting distillation boundaries, and more difficult it is to achieve high-purity products [13]. Consequently, the use of conceptual design principles in simulating and designing a process can be a suitable strategy to overcome the design and operational challenges and select the optimal scenario for separation. Applying the appropriate shortcut method for conceptual design plays a key role in the precise determination of the characteristics of non-ideal separation units [28,29]. In this study, the Extended boundary value shortcut method (EBVSM) is used to examine the feasibility of separation and determine the exact value of design and operational variables that affect the investment costs and energy of a three-phase distillation unit (such as the number of stages, feed stage, reflux ratio, condenser and reboiler duty, operating pressure, etc.).

The EBVSM calculations are based on mass and enthalpy balances and the thermodynamic equilibrium law in the enrichment and stripping sections of the column. In this method, the calculations of the components' concentration and enthalpies of liquid and vapor flow with phase equilibrium calculations are performed tray to tray for both refining and stripping sections starting at both ends of the column. Separation of components is practically feasible when the concentration profiles of the components in the enrichment and stripping sections intersect [8,11]. These features make the EBVSM a powerful tool for the conceptual design of non-ideal columns.

EBVSM was implemented in the powerful Aspen Plus software environment (version 2019). Using this method, the design of the column was performed precisely by identifying the purity or recovery of the products, the reflux ratio, and the operating pressure for a specific feed. According to the number of stages and condenser and reboiler duties, the performance of the three-phase distillation unit was evaluated.

In order to predict the equilibrium and thermodynamic analysis of this polar and highly non-ideal mixture, the NRTL activity model was used in the Aspen Plus software environment. The binary interaction parameters for this model were taken from Aspen Plus database. Initially, the NRTL model predictions were validated with the binary experimental equilibrium data (ethanol/water, ethyl acetate/water, and ethanol/ethyl acetate). The predictions were in good agreement with the experimental data. For brevity, these results were not presented. The ternary diagram of the ethyl acetate/water/ethanol mixture along with the boundaries and distillation regions at 1 atm pressure is shown in Fig. 2. As it is known, this mixture has a ternary azeotrope[1] and three binary azeotropes[2] (all homogeneous and of minimum boiling type).

With regard to the distillation boundaries shown in Fig. 2, three distillation regions are visible. The stable components in the first to third distillation regions are respectively ethyl acetate, ethanol and water, and therefore, they are considered as the main components in the bottom's product of the column. The formation of azeotropic points and multiple distillation regions can complicate the process of separating the mixture. In order to comprehensively evaluate the performance of the separation column, different feeds were considered in each region. As shown in Fig. 2, the place of the intended feeds is on the mass balance line leading to the stable node of each region. Subsequently, the conceptual design of the three-phase column for the mentioned feeds was performed to determine the best region

[1] Composition: 28.17 mol% H_2O, 18.12 mol% ethanol and 53.71 mol% ethyl acetate
[2] Composition: (89.52 mol% ethanol and 10.48 mol% H_2O), (67.34 mol% ethyl acetate and 32.66 mol% H_2O), (55.33 mol% ethyl acetate and 44.67 mol% ethanol)

for the distillation process. For correct comparison, all designs were performed at 1 atm operating pressure and a reflux ratio of 1.5 for 100 kmolF/hr feeds at a temperature of 70˚C.

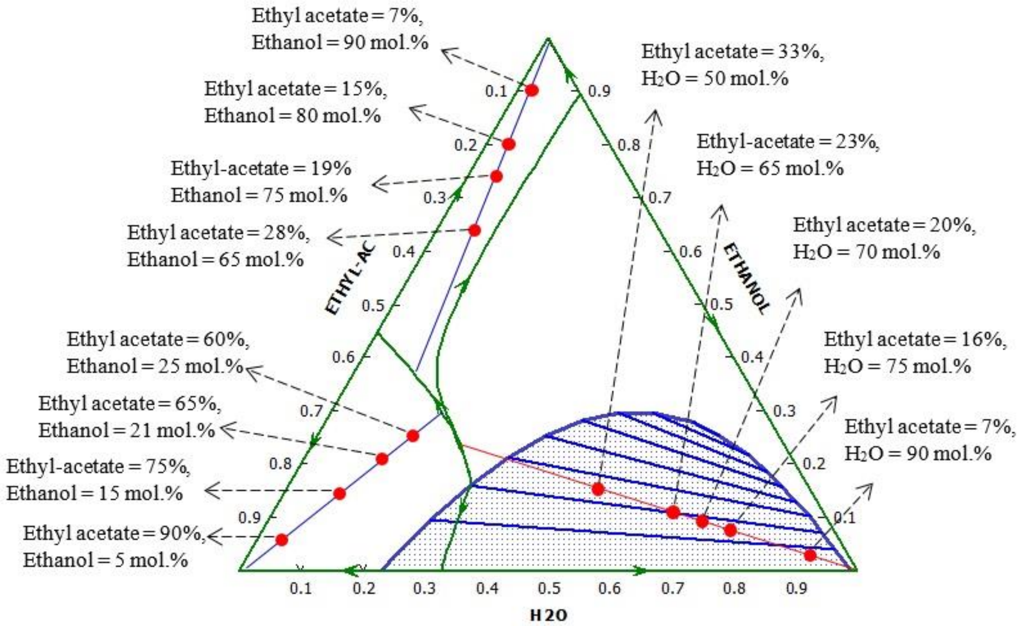

Fig. 2. Ternary diagram of ethyl acetate/water/ethanol with azeotropic points, boundaries, distillation region and the intended feed locations

It should be noted that in each region, conceptual design was carried out at different recovery values than that of the stable component of that region, and the possibility of separation was evaluated. Finally, the results related to the recovery value were selected to be minimum in terms of the number of the theoretical stages required and duty in the condenser and reboiler and maximum in terms of the purity of the received products. The results of the conceptual design for the feeds included in the three regions at the optimal recovery value of the stable component at the bottom of the column are given in Table 1.

Table 1. Results from conceptual design for different feeds on the mass balance line in the ternary regions

Mole percent of feed stable component	Optimal recovery rate from the stable component in the bottom	Stable component purity in bottom (mole %)	Number of stages	Duty (kW)
Region 1				
60	30	84.7	10	3703.67
65	45	91.2	17	3184.59
75	60	95.7	11	2466.59
90	75	98.9	13	1467.22
Region 2				
65	60	91	17	2926.58
75	70	92.5	11	2243.25
80	50	93.5	7	3024.21
90	65	96.6	8	2378.86
Region 3				
50	65	82.2	4	2906.67

By comparing the results presented in Table 1, it should be noted that the second region, in terms of the number of stages required and duty, shows a better situation than the other regions, which reflects the ease of the separation in this region. Only, the purity of the received product in this region is slightly lower than the first one. This drawback is due to the small extent of the second distillation region, which somewhat lowers the region's flexibility to perform the separation process. In fact, with a slight change in the operating pressure of the process (as assessed in the next section), this problem can be easily resolved.

Another advantage of the second region is that by separating ethanol from the bottom of the tower, the overhead products include ethyl acetate and water, which, due to the low miscibility of these two components, can be easily separated in the decanter. This separation potential cannot be achieved in other regions.

Finally, based on the three indices, purity, the number of stages and the duty, it can be concluded that the second region is the best region for performing the three-phase distillation of the desired mixture. Therefore, the main focus of this study was on the second distillation region, and the effect of pressure on the separation flexibility was studied for feeds with a concentration of less than 80 mol% from the stable component (ethanol). Feeds with a concentration higher than 80 mol% of the stable component were not evaluated because of high initial purity.

3. Results and discussion

In this section, to improve the process performance, the effect of operating pressure and reflux ratio on the conceptual design has been investigated. In the following, based on the results, rigorous simulation and process optimization were performed through sensitivity analysis. Finally, the precise design of the tray and packed three-phase columns was performed.

3.1. Effect of operating pressure

In this section, the effect of pressure variation on the extent of the distillation region, ease of separation, purity of received products and the amount of energy consumption (duty) in the second region (selected from the previous stage) has been evaluated through conceptual design. The design of the azeotropic column was carried out at 0.8, 1, 2, 3, 6 and 8 atm for separating feeds containing 75 mol% of the stable component (ethanol). The reflux ratio was constant and equal to 1.5. The distillation boundaries along with the location and composition of the feeds at each pressure are shown in Fig. 3.

As shown in Fig. 3, at lower pressures, the second distillation region is narrower and thicker and becomes wider and smaller at higher pressures. Due to shrinkage of the second distillation region, it is not possible to perform three-phase distillation at pressures above 8 atm.

The results of conceptual design at various pressures in the optimal recovery amount of stable component at the bottom of the column for each feed were given in Table 2. It should be noted that at 8 atm pressure, only two feeds were considered due to the shrinking of the distillation region. For a feed containing 12 %mol ethyl acetate, conceptual design was not possible at any recovery amount.

At 0.8 atm pressure (and lower pressures), in addition to the costs of creating vacuum conditions, the lack of design flexibility due to the narrowing of the second distillation region is one of the disadvantages of using these operating conditions. At pressures higher than 3 atm, high pressure expenses and shrinkage of the second distillation region (which leads to the achievement of products with low purity) does not justify the application of such operating conditions. In addition, increasing pressure elevates the temperature of mixture bubble and dew points and increases duty of the process.

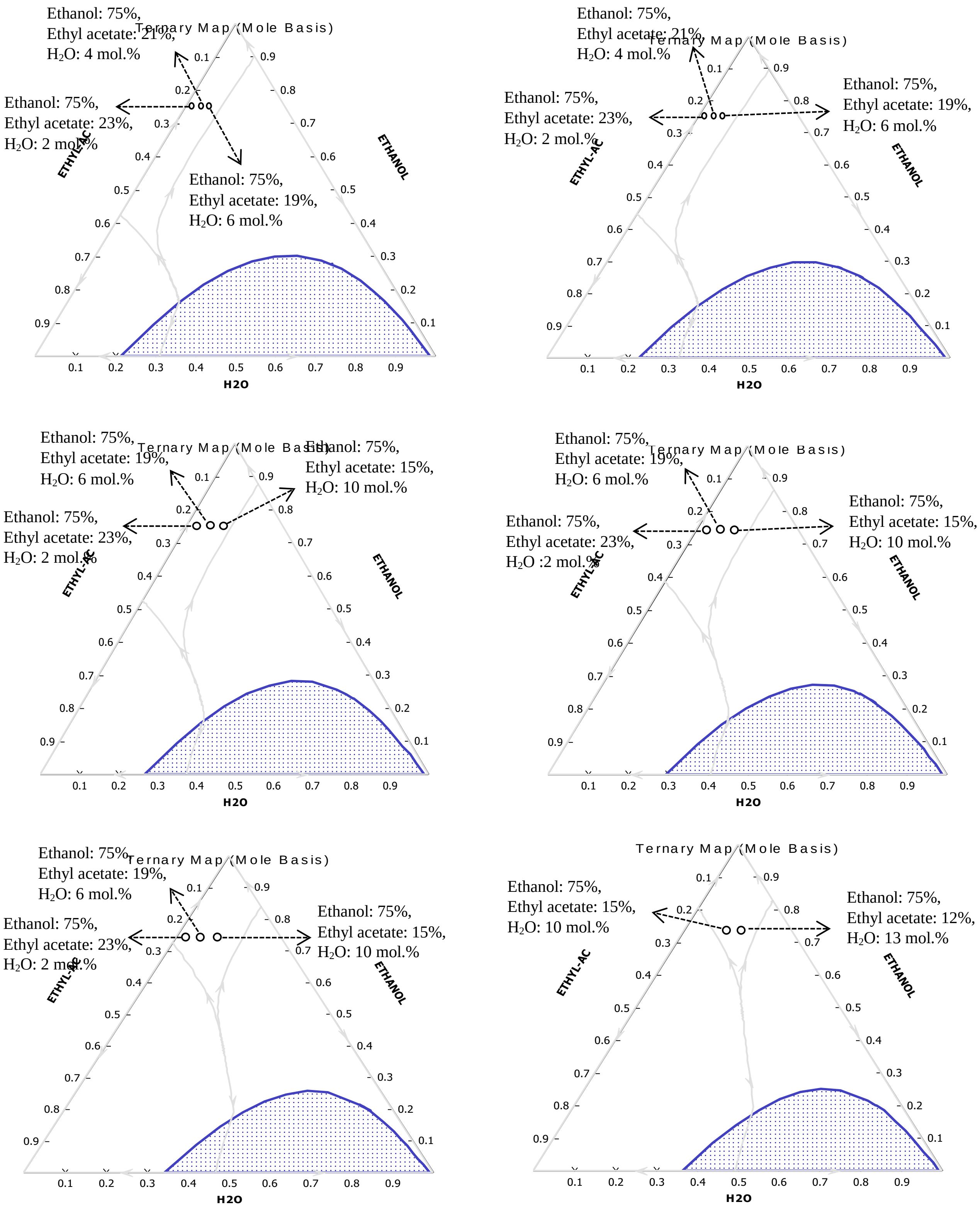

Fig. 3. The distillation boundaries and the composition of the feeds included in the second distillation region at different pressures: 0.8 atm (upper left), 1 atm (upper right), 2 atm (center left), 3 atm (center right), 6 atm (bottom left), 8 atm (bottom right)

Comparing the results presented in Table 2, it is clear that the highest purity (96.2 mol% ethanol) was obtained at the operating pressure of 1 atm for a feed containing 75% ethanol, 23% ethyl acetate, and 2% mol water. Conceptual design for other feeds at this pressure has also resulted in a purity above 90%. The reason for this is the greater extent of the second distillation region at this pressure compared to other operating pressures, which leads to increased flexibility of the separation process of the azeotropic mixture. As a result, 1 atm is introduced as the optimal operating pressure for a three-phase distillation process. However, the number of stages required in this operating pressure is somewhat high, but it is easy to overcome this problem by changing the reflux ratio (evaluated in the next section).

Table 2. Results of conceptual design at various pressures

Operating pressure (atm)	Molar percentage composition of the feed	Ethanol purity in the bottom (% mole)	Number of stages	Duty (kW)
0.8	75% ethanol, 19% ethyl acetate, 6% water	90.1	4	2885.538
	75% ethanol, 21% ethyl acetate, 4% water	93.2	7	1584.219
	75% ethanol, 23% ethyl acetate, 2% water	94.0	11	2696.22
1	75% ethanol, 19% ethyl acetate, 6% water	92.5	11	2243.25
	75% ethanol, 21% ethyl acetate, 4% water	94.5	14	1606.179
	75% ethanol, 23% ethyl acetate, 2% water	96.2	18	2335.78
2	75% ethanol, 19% ethyl acetate, 6% water	93.1	18	2888.62
	75% ethanol, 21% ethyl acetate, 4% water	94.8	17	2915.7
	75% ethanol, 23% ethyl acetate, 2% water	96.1	18	2933.42
3	75% ethanol, 15% ethyl acetate, 10% water	87.0	12	3170.47
	75% ethanol, 19% ethyl acetate, 6% water	93.8	23	3088.14
	75% ethanol, 23% ethyl acetate, 2% water	96.1	30	3301.8
6	75% ethanol, 15% ethyl acetate, 10% water	89.7	25	3544.43
	75% ethanol, 19% ethyl acetate, 6% water	86.7	13	3638.99
	75% ethanol, 23% ethyl acetate, 2% water	78.8	7	3461.91
8	75% ethanol, 12% ethyl acetate, 13% water	-	-	-
	75% ethanol, 15% ethyl acetate, 10% water	83.3	22	3360.22

3.2. Effect of reflux ratio (R) at optimal pressure

In this section, the effect of R on conceptual design in optimal operating pressure has been investigated. Fig. 4 shows the effect of R on the number of equilibrium stages and duty at 1 atm pressure. Obviously, with increasing R, the number of equilibrium stages decreases and duty increases. Initially, with increasing R from 1.4 to 2, a sharp reduction in the number of equilibrium stages occurs and reaches 13 from 25, but after that, the process of reducing the number of stages occurs with a gentle slope.

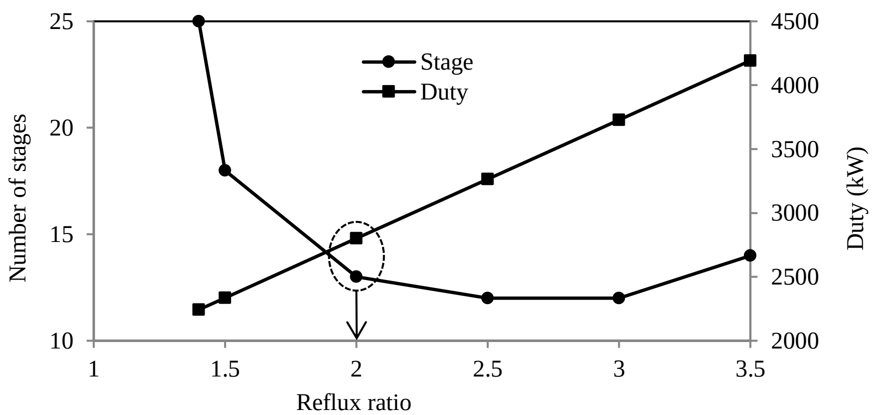

Fig. 4. Effect of reflux ratio on the number of stages and duty for a feed containing 75% ethanol, 23% ethyl acetate, and 2%mol water at 1atm

It is clear from Fig. 4 that the minimum R value obtained at 1 atm pressure which leads to a possible design is 1.4. Regarding the general principle of $R_{opt} = (1.2\text{-}2)\times R_{min}$ and the profiles presented in Fig. 4, a value should be considered as the optimal R, in which the number of required stages and duty are minimal. Therefore, the reflux ratio of 2 is desirable in this regard. The results of the conceptual design in the optimum R value are given in Table 3. It should be noted that by changing the reflux ratio, there was no significant change in the purity of the products. Now, based on the results presented in Table 3, the rigorous simulation of the three-phase distillation column is performed, which is discussed in the next section.

Table 3. The results of conceptual design in the optimal reflux ratio for a feed containing 75% ethanol, 23% ethyl acetate, and 2%mol water at 1atm

Reflux flow	Number of stages	Feed stage	Duty (kW)	The composition of the components in the bottom (mol%)		
2	13	7	2799.97	96.2 (ethanol)	2.1 (ethyl acetate)	1.6 (H_2O)

3.3. Rigorous simulation

The rigorous simulation of the three-phase columns has always been a challenging issue due to the complexity of the convergence caused by the severe non-ideality of the system. In this research, using a suitable conceptual design method as the base platform to simulate the three-phase column, these problems have been overcome. *Radfrac* model in the Aspen Plus environment with 13 equilibrium stages was used for the precise simulation. This model can be simulated both equilibrium-based and rate-based (based on mass transfer). Since the rate-based model is implemented only for two-phase vapor-liquid processes, the simulation was equilibrium-based in this study.

3.4. Sensitivity analysis

In this section, the effect of the important operating and design variables on the performance of the three-phase column was determined by sensitivity analysis in the Aspen Plus environment. Using the sensitivity analysis and taking into account different objective functions, the optimal range of process variables was determined. Reducing process duty and increasing bottoms purity were considered as two objective functions in the sensitivity analysis. In order to achieve these objective functions, the operational or continuous variables (such as reflux ratio, feed temperature, and bottom flowrate) and design variables or discrete ones (such as the number of equilibrium stages and feed tray) are altered within the possible range (in accordance with Table 4).

Table 4. Range of variations of operational (continuous) and design (discrete) process variables along with the initial value of each variable in the sensitivity analysis separately

Variable	Reflux ratio	Number of stages	Feed stage	Feed temperature (°C)	Bottom flowrate (kmol/hr)
Range of changes	0.5-10	6-23	2-13	66-80	35-80
The initial value in distinct sensitivity analysis	2	13	7	72	55

Initially, sensitivity analysis was performed distinctly to evaluate the effect of each variable (other variables were considered constant). After determining the optimal range of each variable, an integrated sensitivity analysis was performed to evaluate the impact of all the variables simultaneously.

3.4.1. Effect of reflux ratio

The effect of R on process duty and bottom composition are shown in Fig. 5. As can be seen, with increasing R, duty and ethanol purity at the bottom of the column are enhanced. However, at R values higher than 5, the purity of ethanol increases with a very mild slope, such that it can be considered almost constant. As a result, as shown in Fig. 5, the optimum R range was considered to be between 2 and 5.

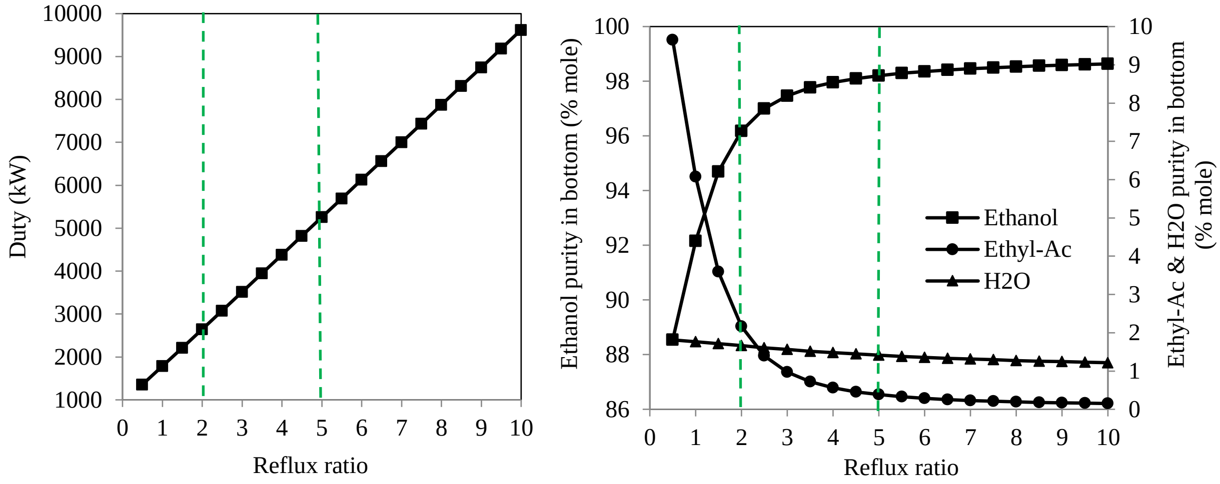

Fig. 5. The effect of R on duty (left) and the bottom composition (right)

3.4.2. The effect of the number of stages

The effects of the number of equilibrium stages on process duty and bottom composition are shown in Fig. 6. As it is expected, increasing the number of stages in the column will increase the purity of ethanol in the bottom and reduce duty. It is clear that up to stage 13, the process of duty reduction and ethanol purity increase will occur with a steep slope, while this trend continues with a very gentle slope after stage 19. Thus, as shown in Fig. 6, the number of optimal stages was considered between 13 and 19.

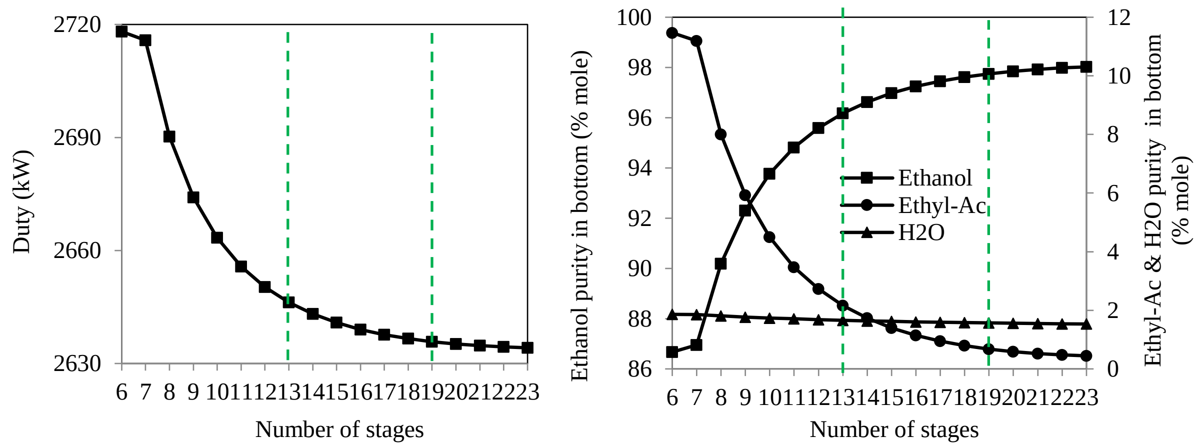

Fig. 6. The effect of the number of stages on duty (left) and the bottom composition (right)

3.4.3. The effect of feed stage

Fig. 7 shows the effect of the feed stage on process duty and the bottom composition. As shown, changing the feed stage does not affect duty, but if the feed is introduced at the mid-column, ethanol purity will increase significantly. As a result, the optimum feed location was considered between stages 4 and 8.

3.4.4. The effect of feed temperature

The effect of feed temperature on process duty and bottom composition is presented in Fig 8. It is obvious that until reaching the temperature of 72°C, duty and purity of the bottom's components are almost constant, but at temperatures 72 to 76°C, a sudden drop in duty and purity of the components occurs. Therefore, with respect to the purity of ethanol and duty, the optimum temperature range was considered between 70 and 74°C.

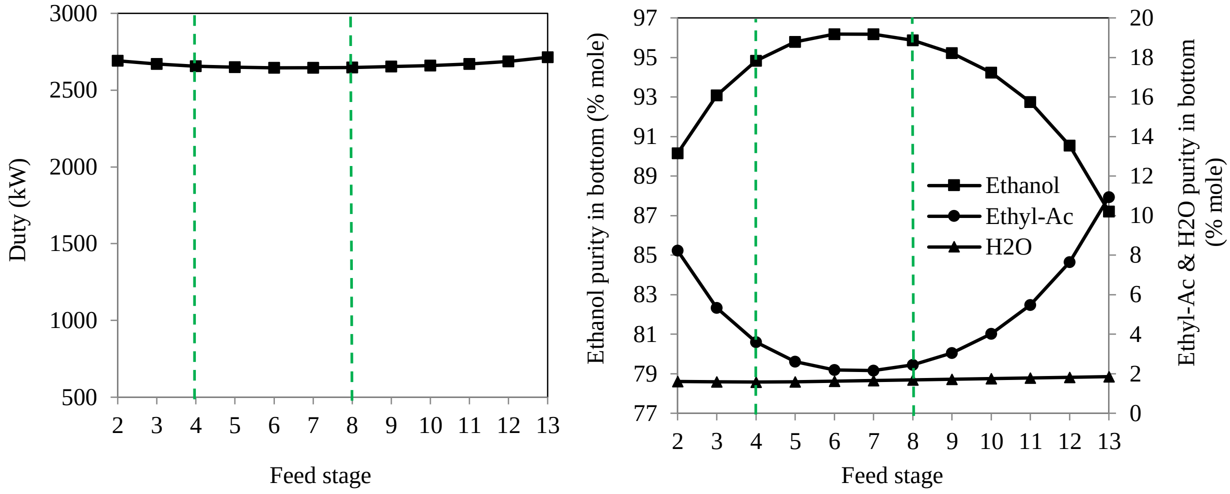

Fig. 7. The effect of feed inlet location on duty (left) and bottom composition (right).

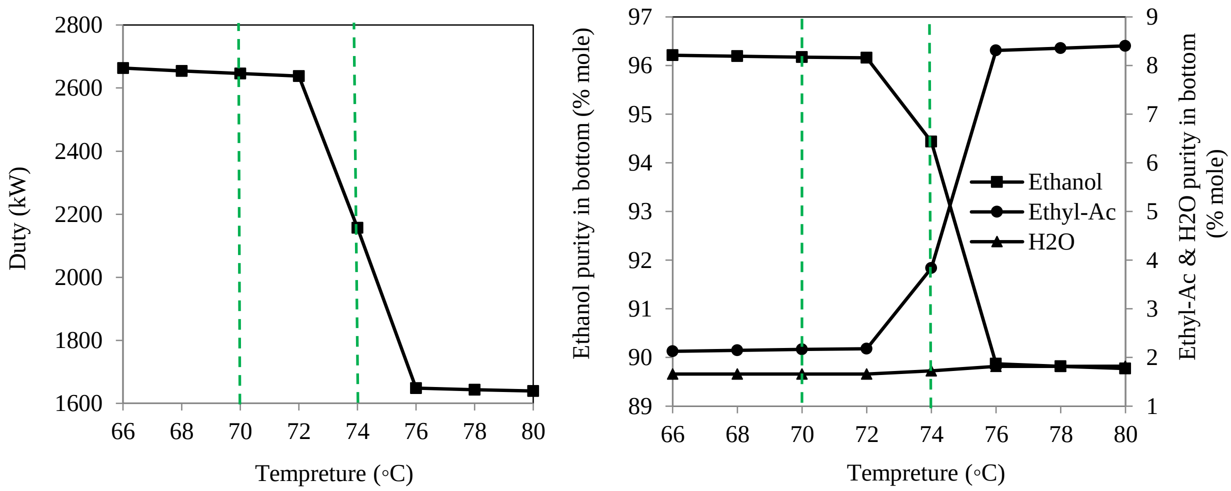

Fig. 8. The effect of feed temperature on duty (left) and bottom composition (right).

3.4.5. The effect of bottom flowrate

In Fig. 9, the effect of bottom flowrate on the process duty and the bottom composition is shown. It is clear that with increasing flowrate at the bottom of the column, ethanol purity and duty decrease. This declining trend is enhanced for ethanol purity at flowrates higher than 55 kmol/hr. As a result, the optimum range for the downstream flowrate was considered between 45 and 60 kmol/hr.

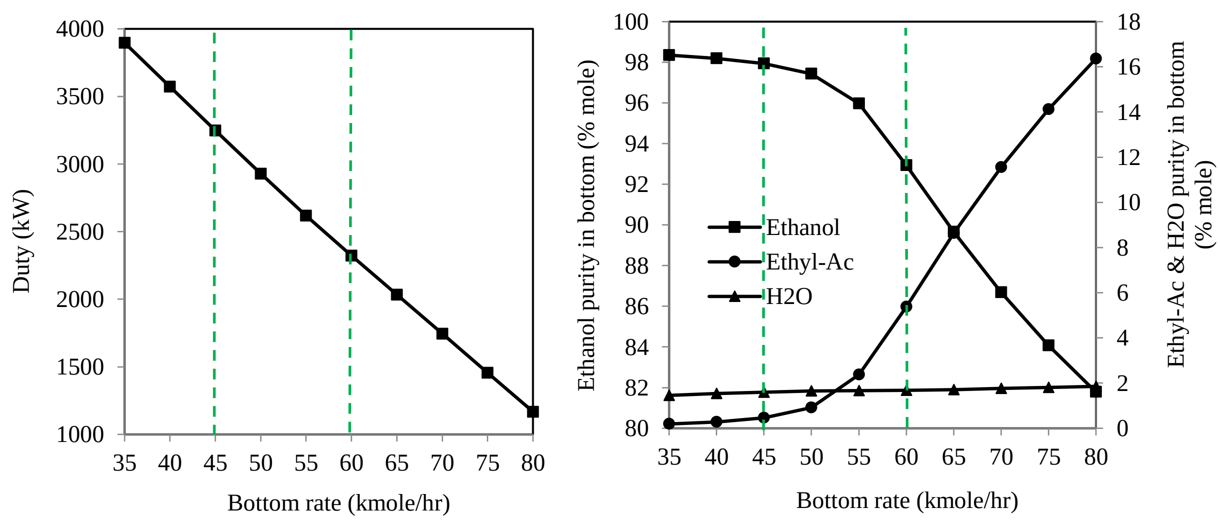

Fig. 9. The effect of bottom flowrate on duty (left) and bottom composition (right)

So far, the optimal range of all process variables has been determined distinctly. Subsequently, by performing simultaneous (integrated) sensitivity analysis on all variables in their optimal ranges, the general optimization of the process was carried out in such a way that the highest ethanol purity was obtained with the lowest duty rate. The final optimal values for the process variables are given in Table 5.

Table 5. Final optimal values of the process operational and design variables

Reflux ratio	Number of stages	Feed stage	Feed temperature (°C)	Flow rate at the bottom of the column (kmol/hr)	Purity of ethanol (% mol)	Purity of ethyl acetate (% mol)	Purity of water (% mol)	Duty rate (kW)
4.5	18	5	72	55	98.70	0.06	1.23	4754.14

As shown in Table 5, performing precise simulation based on conceptual design principles and process optimization through sensitivity analysis led to the achievement of high-purity ethanol (98.7 mol%, 99.5 weight%) with the possible minimum number of stages (18 stages including condenser and reboiler) and duty. In accordance with the international standards (EN 15376, ASTM D 4806), ethanol should have a purity equal to or greater than 98.7 mol% (99.5 weight%) to be used as a raw material in chemical industries or fuel in the current engines [30]. As indicated in Table 5, performing simulation and optimization based on conceptual design principles has led to the achievement of ethanol in accordance with the defined standards.

3.5. Column design

In this section, the rigorous design of the three-phase distillation column was performed based on the results of the optimal model. Since sieve trays and Mellapak packings have known mass transfer relations and are widely used in the interior design of tray and packed distillation columns [31], the design of the three-phase column in this study was based on their use. The rigorous design was performed using the "column sizing tools" in the Aspen Plus environment. Table 6 presents the design characteristics of the columns employed. It should be noted that the efficiency of sieve trays in tray columns was 70% and the height equivalent to a theoretical plate (HETP) was considered to be 0.4 m according to the Sulzer brochure [32, 33].

Table 6. The design characteristics of the tray and packed three-phase columns

Value	Parameter
Sieve tray	
Flooding approach (%)	80
Hole diameter (m)	0.0045
Pitch (m)	0.012 (Triangular)
Hole area/active area	0.127
Number of passes	1
Mellapak packing	
Material	Stainless steel
HETP (m)	0.4
Height (m)	6.4
Surface area (cm^2/cc)	2.56 = 0.0256 m^2/m^3

Tray spacing in the tray column has a significant impact on the performance and dimensions of the column, so determining its optimal value is of particular importance [20]. Fig. 10 shows the effect of the tray spacing on column diameter. Clearly, as tray spacing increases, the diameter required for the column decreases. This decreasing trend is observed to a value of 0.55 m with a relatively steep slope, but then it decreases with a very gentle slope. As a result, the value of 0.55 m was considered as the optimum value of tray spacing. With this optimum value, the diameter and height required for the tray column will be 1.27 m and 8.8 m, respectively.

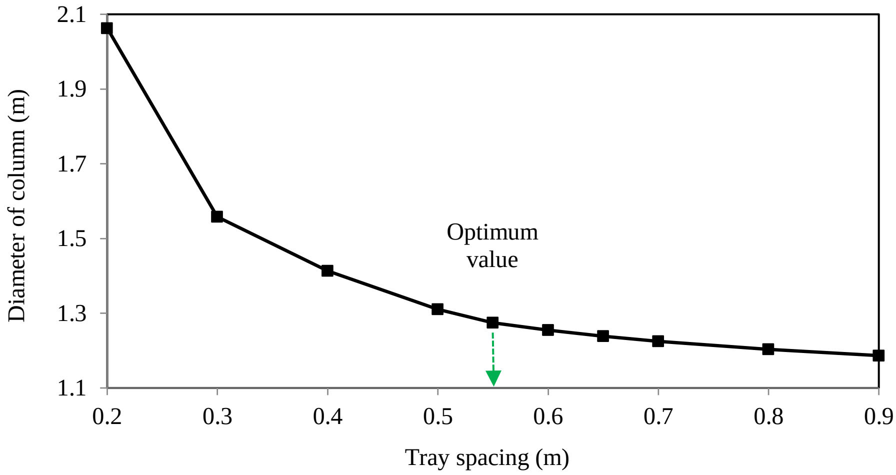

Fig. 10. The effect of tray spacing on tray column diameter

The size of the Mellapak packings has a significant effect on the performance and dimensions of the packed column. Fig. 11 shows the effect of the size of these packings on the diameter of the packed column. As it is known, the use of Mellapak 64x for this process leads to the lowest diameter of the column (1.03 m). With this diameter, the height required for the column was obtained 4.6 m. Therefore, the use of this type of Mellapak packing in the structure of the column is recommended.

By comparing the size required for the tray and packed columns, it is determined that packed columns are smaller in size than tray columns. Thus, the use of packed column for the three-phase distillation process of ethyl acetate reduces investment costs.

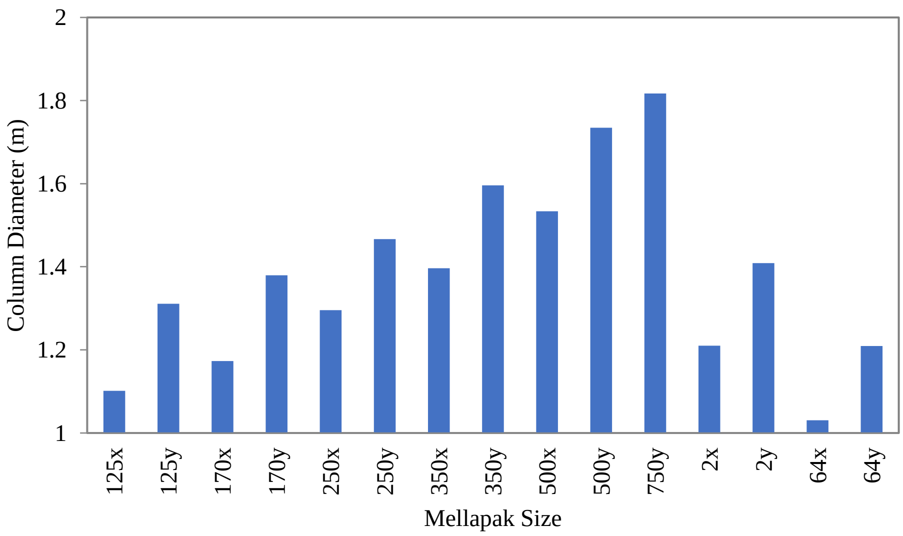

Fig. 11. The effect of the size of the Mellapak packing on the diameter of the packed column

4. Conclusion

Deethanolization in the ethyl acetate process is a challenging and costly problem. In this research, the three-phase distillation method was used for deethanolization based on the principle of creating heterogeneity in the liquid phase and minimizing the boiling point of the mixture. In order to select the best deethanolization scenario from the non-ideal water/ethanol/ethyl acetate mixture, the conceptual design of the three-phase column in different parts of the ternary distillation regions was performed based on the extended boundary value shortcut method.

Based on the results and considering the three indices of product purity, the number of stages and duty, it was determined that the best operating region of the three-phase unit is the second distillation region with ethanol as the stable component. Then, in order to increase the flexibility of the separation process in the second distillation region, the conceptual design of the column was carried out at different operating pressures and reflux ratios. It was determined that performing the process at 1 atm and R = 2 resulted in the highest purity and lowest duty.

Subsequently, based on the conceptual design results, the simulation and optimization of the three-phase distillation unit was performed by sensitivity analysis distinctly and simultaneously on the operating and design variables, and ethanol was separated using 18 equilibrium stages with the purity of 98.7 mol% and duty of 4754.14 kW.

Finally, with the rigorous design of the three-phase packed column using Mellapak 64x packing, the diameter and height required for the column were 1.03 m and 4.6 m, respectively, which is lower than the tray column with sieve trays.

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