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Distillation plant performance implications of high distillate-purity during ethanol-water pre-concentration

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Abstract

This paper presents the effect of distillate purity on the performance of ethanol-water distillation column before azeotrope. The factors affected include the required number of stages, reflux ratio, reboiler and condenser duties, investigated using different feed thermal conditions. The model was set using Aspen Plus® V10 simulation software and used to establish the impact of operating conditions due to high distillate purity. The aim was to increase distillate purity while minimizing energy consumption. The research highlights the importance of modeling and simulation for optimizing distillation processes to reduce energy consumption. The feed flow rate was 1000 kmol/h, with composition ranging from 0.1 to 0.6 mol/mol. The number of stages varied from 8 to 20 while the reflux ratio ranged between 1 and 8. The required number of stages, reflux ratio, condenser and reboiler duties increased with increasing desired distillate purity. The reboiler and condenser duties increased with distillate purity according to 3rd order polynomial functions. It was concluded that the pre-concentration column can concentrate dilute solutions to higher purity closer to the azeotropic point. Further studies were recommended on the non-linear behavior and large energy consumption.

Keywords: Ethanol-water mixtures, distillate purity, reflux ratio, condenser and reboiler duties, modeling and simulation, feed thermal conditions.

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1. Introduction

1.1. Global demand for high-purity ethanol

The global demand for high-purity ethanol, particularly for fuel and industrial applications, has been steadily increasing, with projections indicating significant growth. As of 2023, the fuel ethanol market alone is expected to surpass 35 billion gallons annually due to policies promoting renewable energy and the rising use of ethanol as a biofuel additive (Market Research Future, 2023). The industrial ethanol market, including pharmaceutical and chemical uses, also shows robust demand, driven by stringent purity requirements (Grand View Research, 2022). This trend underscores the necessity for efficient production methods to meet high-purity standards.

1.2. The driving force for high-purity ethanol

Distillation is a technique used in the chemical industry to separate and purify liquid mixtures with appreciable differences in boiling points, such as ethanol-water mixtures, reported in this study. The necessity for high distillate purity in ethanol-water distillation below the azeotropic point is driven by the requirements of various applications, particularly in the fuel sector (US-DOE, 2020), medical applications (WHO, 2020) and industrial sectors (Klein and Klein, 2011; Gerbaud *et al.*, 2018). Achieving this purity involves overcoming significant technical and economic challenges, necessitating advanced separation techniques and continuous innovations (Kiss and Ignat, 2012). The benefits of high-purity ethanol, which includes improved fuel performance, compliance with industrial standards and higher market value (RFA, 2020), underscore the importance of these efforts. This research, therefore, aims to investigate the implications of high-distillate purity demand on the operational variables during normal distillation of ethanol-water mixtures of low and medium ethanol content (that is, pre-concentration). Innovations in distillation technology, such as the use of diving-wall columns and advanced control systems aim at enhancing separation efficiency and reduce energy consumption (Luyben, 2005; Kiss and Ignat, 2012). Continued research and technological advancements are essential to optimize the production process, making high-purity ethanol more accessible and sustainable.

1.3. Challenges in attaining high-purity ethanol during normal distillation

Ethanol-water distillation below the azeotropic point (approximately 89.5% mol ethanol/mol) is constrained by several technical and economic factors (Kiss and Ignat, 2012). The formation of an azeotrope limits the maximum achievable ethanol concentration via normal distillation due to the identical vapor and liquid composition at this point, necessitating additional separation techniques for higher-purity ethanol (Al-Malah *et al.*, 2019). As the concentration of ethanol increases, the relative volatility between ethanol and water decreases, making the separation process increasingly inefficient and requiring higher reflux ratios, energy inputs and slow product withdrawal rate (Ramirez-Corona *et al.*, 2016; Saifuddin and Sirkar, 2003). This leads to significant energy consumption, especially near the azeotropic concentration, as more stages and energy are needed to achieve further separation (Luyben, 2012). Effective separation below the azeotropic point demand precisely designed distillation columns with numerous theoretical plates or packing materials to maximize contact between the vapor and liquid phases, which complicates equipment design and increases operational costs (Moradi *et al.*, 2014; Li *et al.*, 2016; Hassenrück *et al.*, 2017; Wang *et al.*, 2019; Zhang *et al.*, 2019; Shukla *et al.*, 2019). Furthermore, the operational complexity increases as optimal control of temperature gradients, pressure and reflux ratio is necessary to maintain efficiency and avoid process deviations ((Górak *et al.*, 2018; Huang *et al.*, 2019). These factors collectively make ethanol-water distillation below azeotropic and energy intensive and costly process, necessitating intensive research, sophisticated engineering solutions, software implementation for modeling and simulation and skilled operators to manage the delicate balance between operational parameters (Kiss and Ignat, 2012).

Ethanol-water mixtures are often distilled to separate ethanol from water to obtain a solution of a desired concentration or purity. The azeotropic point, denoted as A_z on the *y-x* diagram (Figure 1), is critical in the separation of ethanol-water mixtures because it marks the end-point for normal distillation. After all, further separation by simple distillation is difficult since the number of stages required for efficient separation increased faster (Wang and Huang, 2020). As shown in Figure 1, *y* and *x* represent the concentration of ethanol in the vapor and liquid phases, respectively. On the *y-x* diagram (Figure 1), this point is also called pinch-point, which necessitates a large number of stages, but also larger condenser and reboiler duties and higher reflux ratio. The final distillate purity, X_D, depends on the initial composition of the mixture or feed composition (X_F), the number of stages, heat duties and the operating conditions of the distillation process, especially the feed thermal conditions (FTCs) (Wang *et al*., 2020b).

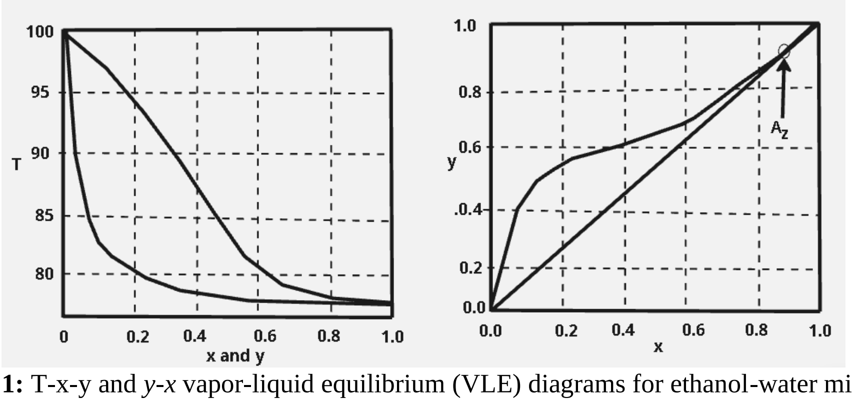


Figure 1: T-x-y and *y-x* vapor-liquid equilibrium (VLE) diagrams for ethanol-water mixtures.

1.4. Vapor-liquid equilibrium curve for ethanol-water mixtures

The ethanol-water mixture is one of the non-ideal mixtures, with a complex vapor-liquid equilibrium curve (as reported in this study. This is because the mixture exhibits azeotrope at 89.5% mol ethanol/mol, as shown in Figure 1. Before the azeotropic point, ethanol and water can be separated by simple distillation aiming at concentrating the solution. The azeotropic mixture is one at which the composition of the vapor produced during distillation is the same as that of the liquid mixture, making further separation impossible by simple distillation (Abe and Kimura, 2017; Inoue and Kaminaka, 2020). To obtain a higher concentration above the azeotropic concentration of ethanol, advanced techniques are employed, including azeotropic distillation or extractive distillation (Xu *et al*., 2019).

Due to the formation of an azeotropic mixture at 89.5 mol% ethanol (or 95.6% ethanol by mass), it is not possible to separate the ethanol-water mixture into ethanol composition above A_z using a normal distillation column (Wang *et al.*, 2020a). In this study, pre-concentration of the dilute ethanol-water mixtures of 0.1 to 0.6 mol/mol was assessed using a normal distillation column to raise the concentration closer to 0.895 mol/mol. Pre-concentration allows for minimal energy consumption (both reboiler and condenser duties) in the azeotropic plant. This paper aims to investigate the effect of the desired distillate purity on the required number of stages, reflux ratio, reboiler and condenser duties in a pre-concentration column.

1.5. The need for modeling and simulation for optimization of distillation process

Because of system complexity, modeling and simulation allow for establishing operating conditions leading to high distillate purity with less energy consumption before real plant operation (Manzo *et al.*, 2012; Mokhtaran *et al.*, 2015; Li *et al.*, 2016; Li *et al.*, 2018). For a feed with low ethanol concentration ($X_F < 0.15$ mol/mol) for instance, modeling for efficient stripping is essential to minimize the loss of ethanol in the bottoms and reduce energy consumption (Liu *et al.*, 2018; Li *et al.*, 2019; Huang *et al.*, 2020). Simulation software like Aspen Plus comprises VLE data and thermodynamic models used to simply the task of estimating model parameters based on the thermodynamic properties of the components of the mixtures to be separated (Tarqvi *et al.*, 2016; Villarrioel *et al.*, 2016; Kulkanrni, 2017; András *et al.*, 2020; Khan and Kumar, 2021).

2. Literature Review

2.1 The factors affected by increasing distillate purity

Several studies have investigated the effect of increasing the product quality (X_D) on various operating parameters for distillation of ethanol-water mixtures before the azeotropic point, including the number of stages required (Khalilpour *et al.*, 2016; Kazemi *et al.*, 2017), the reflux ratio (Manjón *et al.*, 2016; Kongsompong *et al.*, 2018; Wang *et al.*, 2019a), condenser and reboiler duties (Gondal and Hussain, 2017; Kim and Lee, 2018).Wang *et al.* (2019b) investigated how the condenser temperature is affected by increasing X_D on ethanol-water separation by batch distillation. It was reported that high distillate purity requires low condenser temperature and a high reflux ratio. Li *et al.* (2016) investigated the effect of operating pressure on the distillation of ethanol-water mixtures before the azeotropic point. These variables have been studied in detail in this paper. It was reported that increasing the operating pressure increases the number of theoretical plates required for a fixed level of distillate purity. Jalali-Heravi *et al.* (2014) investigated the effect of column diameter on the separation of ethanol-water mixtures. It was reported that increasing the column diameter led to a decrease in the number of theoretical stages required and an increase in the capacity of the column. In this study, operating pressure and column diameter were fixed to reduce the number of variables. Thus, the distillation of ethanol-water mixtures before the azeotropic point can be optimized by carefully controlling various operating parameters to yield the desired distillate purity. Optimization of these parameters should focus on efficient and cost-effective production of high-purity ethanol.

2.2 The effect of the azeotropic point on the required number of stages for ethanol-water mixtures

The required number of stages for the distillation of ethanol-water mixtures before the azeotropic point is significantly influenced by the desired distillate purity. The higher the distillate purity required, the greater the number of stages required. A study by Ramírez-Corona *et al.* (2016) reported that the required number of stages for 95% ethanol distillate purity was around 27, while for 99% purity, it increased to around 40. Similarly, a study by Saifuddin and Sirkar (2003) reported that for a 95%-purity, the required number of stages was 22, while for a 99% purity, it increased to 38 for the ethanol-water system. This can be attributed to the decreased driving force for mass transfer represented by the *y-x* curve being closer to the $y = x$ line, as shown in Figure 1. These are excessively higher numbers of stages, columns of which are expensive to erect since it increases the equipment cost and complexity of the distillation process. Thus, a balance between desired purity and economic feasibility must be considered during the design and operation of an ethanol-water distillation plant. In this study, the number of stages was not allowed to vary but the model was solved for fixed values of $N = 20$, 15, 10 and 8 stages while tracking the distillate purity attained.

2.3 Effect of distillate purity on the required reflux ratio during ethanol-water distillation

The distillate purity affects the required reflux ratio during ethanol-water distillation during pre-concentration. Several studies have investigated this relationship to optimize the separation process and improve the efficiency of ethanol production. The effect of distillate purity on the required reflux ratio and other operating parameters during the distillation of ethanol-water mixtures has been investigated by Seyed-Matin *et al.*, 2012; Hashemi *et al.*, 2017; Olajire and Gbadegesin, 2018), who reported that increasing the distillate purity led to an increase in the required reflux ratio. This implies that the reflux ratio must be increased to obtain a distillate of higher purity. Furthermore, Olajire and Gbadegesin (2018) reported that the optimal reflux ratio could be determined by balancing energy consumption and distillate purity.

2.4 Dependency of condenser duty on distillate purity during ethanol-water distillation

The distillate purity strongly affects the required condenser duty (Li *et al*., 2018). Condenser duty is the amount of heat that must be removed from the vapor leaving at the top of the distillation column to condense it into a liquid product (Seader and Ernest, 2006). The condenser duty is an important parameter that determines the efficiency and operating costs of the distillation process (Wang *et al*., 2019). When the desired purity of the distillate is high, a higher condenser duty is required because of the high volatility of ethanol which restricts condensation, until enough heat is removed (Tang *et al*., 2017; Shukla *et al*., 2019). The thermodynamic data banks show that the heats of condensation for water and ethanol are 40.6 and 38.6 kJ/mol, which must be removed to condense 1 mole of each. High condenser duty is required because ethanol still exists in gaseous form even at low temperatures. Thus, a higher condenser duty allows for more efficient cooling of the vapor. On the other hand, when the desired purity of the distillate is low, a lower condenser duty may be sufficient to achieve efficient separation.

2.5.The effect of distillate purity on reboiler duty during ethanol-water distillation

Reboiler duty is the amount of heat that must be supplied to the bottom part of the column to vaporize the liquid mixture (Luyben, 2005; Seader and Ernest, 2006). It is an important parameter that determines the efficiency of the distillation process and the amount of heating required for effective separation (Hashami *et al*., 2017). When the desired purity of the distillate is high, a higher reboiler duty is required to achieve efficient separation of ethanol and water (Kiss *et al.*, 2015; Mokhtarani *et al*., 2015; Gondal and Hussain, 2017; Zhang *et al*., 2021). This is because a higher reboiler duty allows for more efficient vaporization of the liquid (Jafar *et al*., 2021). Moreover, a higher reflux ratio is required necessitating a large liquid flow rate being refluxed back to the column all of which must be boiled off again. Meanwhile, even the less volatile component must be vaporized to improve rectification and stripping operation in the top and bottom sections, respectively.

Literature suggests that the distillate purity has a significant effect on the required reboiler duty during ethanol-water distillation(Mokhtarani *et al.*, 2015; Saif *et al.*, 2016; Gondal and Hussain, 2017; Zhang, 2018). Studies also highlight the use of various techniques, such as software simulation (Ramirez-Corona *et al.*, 2016; Shukla *et al*., 2019; Andras *et al*., 2020), mixed solvent, and modified columns (Saif *et al*., 2016), to optimize the energy consumption and efficiency of the process. Additionally, some articles use experimental and theoretical approaches to investigate the separation of ethanol-water mixtures and provide insights into the required reboiler duty (Yildrim and Yildrim, 2016). The results show that a higher reboiler duty is required for higher distillate purities.

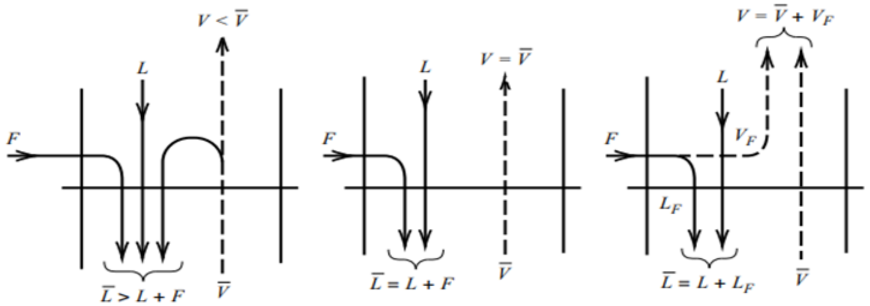
2.6 The effect of feed composition on distillate purity during pre-concentration

The feed composition (X_F) is another important factor in distillation processes which affect distillate purity (Kim *et al*., 2016; Wang *et al*., 2019; Zhang *et al*., 2021). The composition of the feed stream determines how close to the azeotropic point the distillate purity will be, given the number of stages, reflux ratio and column design features (Lee *et al.*, 2018; Li *et al.*, 2019; Doe and Smith, 2020). Therefore, understanding the impact of feed composition on the distillate purity is crucial for optimizing the distillation process. In this paper, the variation of different variables with distillate purity was studied at different fixed values of feed composition was studied. Kim*et al.* (2016) studied the effect of feed composition on the distillate purity for a binary mixture of methanol and water, while Abe and Kimura (2017) and Inoue and Kaminaka (2020) studied the influence of feed composition on the separation of azeotropic mixtures by extractive distillation. Nakamura and Morooka (2019) reported the effect of feed composition on the distillate purity for an isopropanol–water system with entrainers. There have been several studies on the effect of feed composition on distillate purity for ethanol-water mixtures below the azeotropic point, which have shown that the feed composition can significantly affect the purity of the distillate obtained from ethanol-water mixtures below the azeotropic point (Caso and Chen, 2016; Al-Malah *et al.*, 2019; Li *et al.*, 2020).

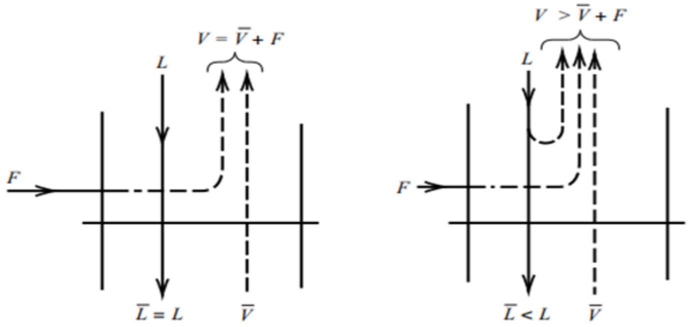
2.7 Effect of feed thermal conditions on the distillate purity for ethanol-water mixtures

Feed thermal conditions (FTCs) refer to the temperature and state of the mixture at which the feed is introduced into a distillation column (Manyele *et al*., 2021), as summarized in Figure 2. The feed temperature can have a significant impact on the efficiency and performance of the distillation process (Albino *et al.*, 2017; Nguyen *et al.*, 2019; Qi *et al.*, 2020; Wang *et al.*, 2020b). When the feed temperature is too high, it can lead to thermal degradation of the components in the feed mixture, which can result in reduced purity of the distillate (Jha *et al.*, 2016; Beshir *et al.*, 2018; Górak *et al.*, 2018; Huang *et al*., 2019). On the other hand, if the feed temperature is too low, it can result in reduced vaporization rates and increased energy consumption. The performance of a distillation process also depends on FTCs. This is because, some FTCs imply feed with high or low heat content reducing or increasing reboiler heat requirements, respectively. Partially vaporized feed (PVF), for instance, allows vapor portion of the feed to enter the rectification section directly and the remaining part (mainly liquid) to travel down the stripping section to the reboiler (Seader and Ernest, 2006). Optimizing the feed thermal conditions is therefore an important consideration in distillation process design and operation. Process control techniques are required in order to attain optimum feed temperature for high performance of the distillation process (Luyben, 2013; Xu *et al.*, 2019).The optimal feed temperature will depend on various factors, such as the composition of the feed mixture, the boiling points of the components, the internal design of the distillation column, and the desired purity of the distillate. In practice, the feed temperature is typically controlled by preheating the feed stream before it enters the distillation column. The preheating temperature can be adjusted to achieve the desired feed temperature and hence optimize the distillation process performance (He and Chen, 2020; Li *et al.*, 2021).

Overall, liquid and vapor flows profile across the column stages provides important visualization on the internal distribution and contact of liquid-vapor phases. This helps in developing flows control strategies in a distillation column so as to achieve the desired separation performance. Extremely high amount of liquid and vapor flows inside the distillation column may cause serious distillation problems such as conning, weeping, flooding, blowing and entraining. These results into increase in column operating expenses, energy requirements and decrease in column separation performance attributed by insufficient contact between liquid and vapor phases flowing counter currently across the column stages.



(a) Sub-cooled liquid feed ($q > 1$) **(b)** Saturated liquid feed ($q = 1$) **(c)** Vapour-liquid mixture feed ($0 < q < 1$)



(d) Saturated vapour feed ($q = 0$) **(e)** Superheated vapour feed ($q < 0$)

Figure 1: Effect of feed thermal conditions on vapor and liquid flows in a distillation column(Green and Perry, 2008).

3. Methodology

3.1 Design considerations for distillation plant used in pre-concentration of ethanol-water mixtures

Designing a distillation plant for ethanol-water mixtures before the azeotropic point requires careful consideration of various design features. Several studies have investigated the optimization of the design features for the efficient and cost-effective production of high-purity ethanol (Luyben, 2013; Seader and Ernest, 2006). Overall, designing an efficient and cost-effective distillation plant for ethanol-water mixtures requires careful consideration of various design features, including tray configuration, reflux and boil-up ratios, and reboiler and condenser design. Optimization of these design features can lead to improved separation efficiency and reduced energy consumption. Table 1 shows the key important points for efficient and cost-effective separation of the water and ethanol.

Table 1.Column design features for ethanol water distillation before azeotropic point.

Column components	Design considerations	Final design	Citations
Column type	The most commonly used column types are packed and tray columns. Packed columns have high efficiency and are less expensive, while tray columns have better turndown ratios and can handle a wider range of operating conditions.	Trayed column	Khalilpour *et al.* (2016); Manyele (2021)
Feed pre-treatment:	The ethanol-water mixture needs to be pre-treated (by filtration or centrifugation) to remove any impurities.	FTC preparation process; filters; heat exchanger	Liu *et al.* (2015)
Tray/column design	The number of trays in the distillation column should be optimized based on the desired purity of the distillate. The use of structured packing and other design features can also improve the efficiency of the column.	10 trays in addition to reboiler and condenser.	Al-Douri *et al.* (2018); Khalilpour *et al.* (2016); Rios-Lopez *et al.* (2019)
Reboiler design	The reboiler should be designed to provide the necessary heat input. The use of plate-type heat exchangers, submerged coils, or other types of heat transfer equipment is critical.	Kettle reboilers	Gondal and Hussain, (2017); Kim and Lee (2018)
Condenser design	The condenser should be designed to efficiently condense the vapor. Shell-and-tube, plate-type or other types of condensers can be used, with appropriate cooling media	Shell and tube condenser	Kim and Lee (2018); Wang *et al.* (2019a)

Reflux Ratio:	The reflux ratio ($R = L/D$) has a significant impact on the separation efficiency and energy consumption. Generally, a higher reflux ratio leads to better separation efficiency but also higher energy consumption.	Wide range: R from 2 to 10	Manjón *et al.* (2016); Caballero and Ruiz,(2018); Olajire *et al.* (2018)
Energy recovery and efficiency	The use of energy recovery systems, such as heat exchangers, can help reduce the overall energy consumption of the distillation plant	Shell and tube heat exchangers for heating and cooling to attain different FTCs.	Mokhtarani *et al.* (2015); Tang *et al.* (2017); Lee *et al.* (2021)
Heat Integration	Heat integration (a key design feature) involves the use of heat exchangers to recover heat from the distillate and the bottoms streams and preheat the feed which reduces the energy consumption.	Heat exchangers aimed at attaining different FTCs.	Tang *et al.* (2017)

3.2 Process description

A simplified process flow diagram (PFD) of a distillation column used in this study is shown in Figure 3. The PFD includes the energy input (Kiss *et al.*, 2015) into FRT in order to attain FBP, SHV or PVF and heat removal to attain FDP. After preparing the selected feed condition, the feed enters the column with properties described by flow rate (F), composition (X_F), molar enthalpy (H_F), and feed temperature (T_F). A section of the feed plate is also shown on the right-hand side, to indicate the details of liquid and vapor flow at the feed plate (Kiss *et al.*, 2015; Manyele, 2021).A feed with composition X_F (mol ethanol/mol) enters in the middle section of the column (at a location or tray denoted as f_s), leading to a product stream with high ethanol content (distillate) at the top of the column and a bottoms stream with very low ethanol concentration, X_B. This study focusses on the factors affected by increasing the distillate purity or distillate purity. The molar flow rates of the streams are denoted as F, D and B, representing the feed, distillate and bottoms streams, respectively. Cooling water was used to condense the vapors via a condenser, while steam was supplied to the reboiler to vaporize the liquid mixture.

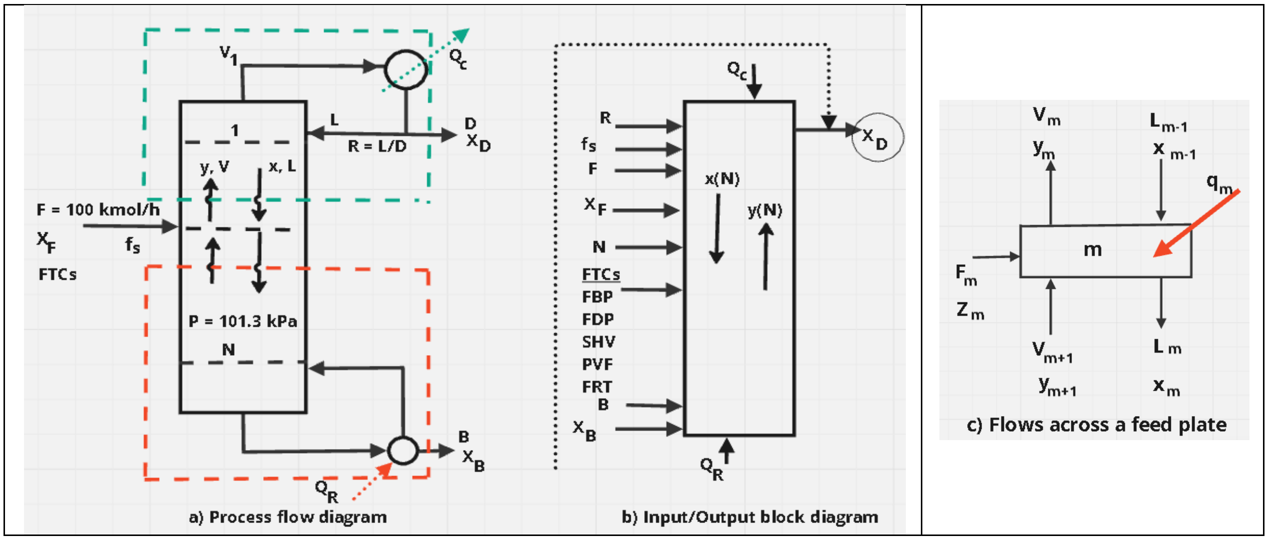


Figure 3: Process flow diagram for the distillation process used in this modeling work (a) PFD; b) Variables affecting X_D; c) Mass and energy balances across the feed plate).

A feed mixture of two components (in this case ethanol and water), was separated into two streams, that is, an overhead distillate with a composition denoted as X_D, and bottoms at a concentration denoted as $X_{B,}$ whose composition differs from that of the feed. The bottoms product is almost always a liquid, but the distillate in this model was either a saturated liquid reflux or liquid distillate at a bubble-point temperature (total condenser). Heat is transferred into the process in the "reboiler" (typically a shell-and-tube heat exchanger, specifically, kettle reboiler) to vaporize some of the liquid from the base of the column. The heat-transfer rate in the reboiler is denoted as Q_r (measured in MW). The vapor coming from the top of the column is liquefied in a condenser, which is another shell-and-tube heat exchanger. The heat was transferred out of the condenser at a rate denoted as Q_c, also measured in MW. Liquid from the condenser drops into the reflux drum, from which, the distillate product is removed. In addition, the reflux is fed back to the top tray of the column. This liquid reflux and the vapor boilup in the base of the column are necessary to achieve the separation or "fractionation" of chemical components, in this case, ethanol and water. At the bottom of the column, a partial re-boiler was used to produce vapor boilup with boil-up ratio V_B=V/B where V is the vapor flow rate from the reboiler to the column bottom and B is the liquid bottoms product withdrawn from the reboiler.

3.3 Mathematical model development and vapor-liquid equilibrium

To analyze the effect of increasing distillate purity, X_D, on the distillation column performance and operational parameters, equilibrium data is required, which is held in software libraries, starting with K-values data for ethanol and water system. The K-value of ethanol, or component *i*, is defined as per Equation (1) (Kister, 1992; Holland, 2011; Anton, 2013):

$$K - value\ (K_i) = \frac{y_i}{x_i} = \gamma_i \psi_i \frac{\phi_i^L}{\phi_i^V} \frac{p_i^o}{p} \tag{1}$$

where ϕ_i^L is the liquid fugacity coefficient that accounts for the effect of vapour non-ideality on liquid fugacity and it is also estimated from equation of state on basis of system temperature and pure component vapor pressure. In Equation (1), γ_i is the liquid activity coefficient that corrects the liquid fugacity for the effect of composition and ψ_i is the pointing correction factor that accounts for the effect of pressure on liquid fugacity, it accounts for the difference between pure component vapour pressure and mixture pressure. In Equation (1), p_i^o is the vapor pressure of component *i* at standard conditions, and *p* is the total pressure.

The relative volatility, defined as the ratio of K-values between the more volatile component to the less volatile component (Kister, 1992)., is given in Equation (2): As highlighted in many literatures higher value of relative volatility is preferred in distillation processes due to the fact that separation process by difference in volatility is easier for higher relative volatility.

$$Relative\ volatility\ (\alpha_{ij}) = \frac{K - value\ component\ i\ (K_i)}{K - value\ component\ j\ (K_j)} = \frac{y_i x_j}{x_i y_j} = \frac{y_i(1 - x_i)}{x_i(1 - y_i)} \tag{2}$$

Therefore, Equation (2) leads to:

$$y_i = \frac{\alpha_{ij} x_i}{1 + (\alpha_{ij} - 1)x_i} \tag{3}$$

For binary system, α_{ij} is taken as simply α, and $x_i = x$ while $y_i = y$, so that Equation (3) becomes:

$$y = \frac{\alpha x}{1 + (\alpha - 1)x} \tag{4}$$

To effectively simulate the distillation process, Aspen Plus develops the molar flows profiles to enable determination of the correct distillation purity at a given values of input variables listed in Figure 3. These profiles depend on bubble and dew point temperatures (Kister, 1992; Holland, 2011; Seader *et al.*, 2006; Anton, 2013), which are obtained when Equations (5) and (6) are satisfied, respectively:

$$\sum_{i=1}^{N_c} y_i = \sum_{i=1}^{N_c} K_i x_i = 1.0 \tag{5}$$

and

$$\sum_{i=1}^{N_c} x_i = \sum_{i=1}^{N_c} \frac{K_i}{y_i} = 1.0 \tag{6}$$

To effectively simulate the flow dynamics of the distillation column, given the FTC (Green and Perry, 2008), Aspen Plus generates among other profiles, the molar enthalpy profiles (Mustapha *et al.*, 2007; Demire, 2013; Adesina and Popoola, 2016; Madyshev *et al.*, 2022) allowing tray modeling by taking into material, equilibrium, summation and heat balances. Also, Aspen Plus determines the values of *y* and *x* across the distillation column, using stage-by-stage calculations, given the inputs listed in Figure 3. The value of *y* for the vapor leaving at the top of the column is taken as the distillate purity, X_D, when a total condenser is used. The mass balance across the top section of the column (above feed plate) leads to the rectifying operating line (ROL) equation, while the bottom section mass balance leads to stripping operating line (SOL) equation (Manyele, 2021).

Equations (7) and (8) present the total flow and component material balances across a feed stage, $m = f_s$, respectively, as per nomenclature presented in Figure 3:

$$F_m + V_{m+1} + L_{m-1} = V_m + L_m \tag{7}$$

$$F_m z_m + V_{m+1} y_{m+1} + L_{m-1} x_{m-1} = V_m y_m + L_m x_m \tag{8}$$

where F_m = feed flow rate into the stage, V_m = vapor flow rate from the stage, L_m = liquid flow rate from the stage, V_{m+1} = vapor flow rate into the stage from the stage below m, L_{m-1} = liquid flow rate into the stage from the stage above m, q_m = heat flow rate into or out the stage. Equation (9), on the other hand, gives the energy balance:

$$F_m H_F + V_{m+1} H^V{}_{m+1} + L_{m-1} H^L{}_{m-1} + q_m = V_m H^V{}_m + L_m H^L{}_m \tag{9}$$

with H_F, H^V and H^L representing enthalpy per unit mass of the feed, vapor and liquid streams, respectively.

3.4 Preparation of the feed to different thermal conditions

At the feed-stage pressure, five feed thermal conditions studied were: feed at room temperature (FRT), (also called sub-cooled liquid depending on temperature), partially vaporized feed (PVF), feed at its boiling point (FBP), feed as superheated vapor (SHV) and feed at dew-point (FDP). The preparation techniques and q-values for the FTCs are summarized in Table 2, which refers also to Figure 3. The feed at room temperature (FRT) do not require preparation, it is fed directly into the column.

Table 2: Methods for preparation of different feed thermal conditions with the range of q-values.

FTC	Preparation method	q value	Temperature (°C)
FRT	No preparation was needed beside maintaining feed concentration	q > 1	30
FBP	1. Determine the boiling point of the feed mixture a distillation curve or a thermodynamic calculation. 2. Heat the feed mixture to its boiling point using a pre-heater or heat exchanging with bottoms. 3. Ensure homogeneous mixing to ensure that the constant composition throughout the feed stream.	0	86.5
PVF	1. Determine the vapor fraction to ensure that the correct amount of liquid and vapor is entering the distillation column (50% vapor) or measured using a vapor-liquid equilibrium (VLE) experiment. 1. Adjusting the feed temperature, pressure, or flow rate to maintain a stable distillation process. 2. Control the vapor-liquid ratio by adjusting the reflux rate or the reboiler duty. 3. The PVF should be well-mixed to ensure homogeneity throughout the feed stream.	0 < q < 1	95.1
FDP	1. Determine the dew point of the feed mixture using a thermodynamic calculation or by experimentally by measuring the temperature at which the first droplets of condensate form from the vapor. 2. Cool the feed mixture to its dew point using a heat exchanger or a refrigeration system. 3. Ensure homogeneous mixing throughout the feed stream.	1	97.3
SHV	1. Determine the feed conditions including the temperature and pressure, to ensure that the feed mixture is completely vaporized. 2. Flashing the feed (below its vapor pressure) can also be alternatively used to completely vaporize the feed 3. Heat the feed mixture to ensure complete vaporization, and heat further to raise degrees of superheat (DSH). This can be done using a reboiler or an external heat source.	< 0	120

3.5 Distillation process model operating conditions

Manyele (2021) presented the mathematical relationships for modeling a binary distillation system, covering equilibrium and mass balance relationships, feed thermal conditions identifier (the *q*-values), condenser and reboiler duties based on energy balances and steam and cooling water mass flow rates for the benzene-toluene mixture. Benzene and toluene, however, form an ideal mixture, without A_z. The equations developed for benzene-toluene mixture can be applied to ethanol-water mixtures, for building an understanding of the process. Differences in relative volatility, boiling points and molar latent heat are then taken care by the model or simulation package upon selection of the components of the mixture. The overall molar composition of the feed with respect to the light component (ethanol), denoted by X_F, was varied between 0.1 and 0.6 mol/mol, to establish how close the distillate can approach the azeotropic molar concentration of ethanol. This forms the focus of this study, that is, to determine the necessary levels of different variables needed to reach higher distillate purity, X_D. The operating conditions used to set up the model in Aspen Plus are shown in Table 3.

Table 3. Operating conditions for the distillation column (at atmospheric pressure, 101.325 kPa).

Parameter	Value or range
Feed flow rate, kmol/h	1,000
Lighter key component	Ethanol, C_2H_5OH
Heavier key component	Water, H_2O
Number of stages, N	8 - 20
Reflux ratio, R	1 - 16
Mole fraction of the lighter component inthe feed, X_F, mol/mol	0.1 – 0.6
Condenser cooling water temperature (in), T_{in}, °C	28
Condenser cooling water temperature (out), T_{out}, °C	75

3.6 Pre-concentration distillation plant model build-up and convergence

The simulation model for the pre-concentration distillation plant was created by using Aspen Plus® V10 simulation software. The first step taken in the model build-up and convergence was property analysis and specifications, that is, component specification, thermodynamic fluid package selection and distillation synthesis analysis.

1) The main components of the distillation process streams were specified (ethanol and water) by stating component identification, type (pure and of conventional type), component name and formula, to accurately determine the appropriate thermodynamic fluid package to be used.

2) The Non-Random Two Liquid-Redlich Kwong (NRTL-RK) thermodynamic model was selected as the base method in this study because it is highly recommended for non-ideal chemical systems (i.e., ethanol-water systems) and it also describes vapor-liquid and liquid-liquid equilibrium data appropriately (Taqvi *et al.*, 2016).

3) Process flowsheet development and simulation which include defining the distillation process connectivity by placing the required unit operations (blocks) from the model library and their respective connecting process streams. This step also included specifying input process stream properties specifications (i.e., pressure, temperature, composition and flow rate), selecting of flowsheet convergence criterion from the convergence block and configuring the flowsheet balance model to solve process convergence problems.

4) Therefore, after solving the process convergence problem modeling and simulation of the converged distillation process was conducted. This was achieved by varying plant operating parameters listed in Figure 3 (input/output block diagram). The results obtained during the distillation process modelling and simulation were then collected and analyzed in MS Excel 2007.

4. Results and Discussion

4.1 Effect of Distillate Purity on Required Number of Stages

To explore the impact of increasing distillate purity during the pre-concentration phase for ethanol-water mixtures, feed composition and reflux ratio were maintained constant. Figure 4 shows the effect of increasing X_D on the number of the theoretical stages N at X_F = 0.6 mol/mol and R = 3, 4 and 5 for a selected FTC, that is, FBP. The number of stages increased from 8 to 20 when X_D was increased from 0.81 to 0.86 mol/mol. In each case, a sharp increase in the number of stages was observed, being highest at a low reflux ratio, R = 3 than at the highest reflux ratio, R = 5, for a given distillate purity. This behavior was observed for all FTCs. The fast increase in the number of stages as X_D increases indicates a large demand on capital cost for the construction of a distillation column with a large number of trays to achieve the desired separation. As shown in Figure 4, a column with a fixed number of trays, say N = 10, can only reach X_D = 0.821, 0.828 and 0.834 mol/mol at fixed reflux ratios of 3, 4 and 5, respectively, when a feed composition of 0.6 mol/mol is used. At N = 20 stages, the corresponding values of X_D were 0.846, 0.854 and 0.860 mol/mol, respectively. There is a small increase in X_D compared to the large increase in the number of stages, showing a greater resistance towards high distillate purity. This is attributable to the formation of azeotrope.

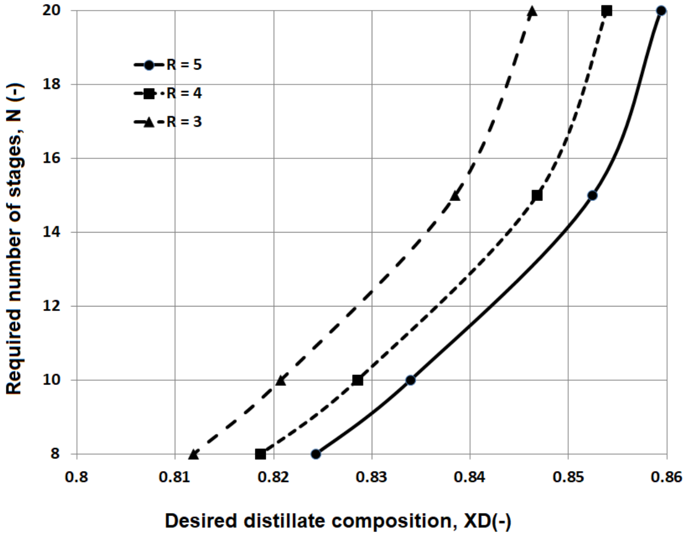


Figure 4: Effect of desired distillate purity on the required number of stages to achieve separation for the FBP at X_F = 0.6 mol/mol, and at constant reflux ratios.

Figure 5 compares the variation of N with X_D for different fixed values of X_F and R and also between FRT, FBP and FDP. In all cases, the number of stages increases faster with distillate purity as shown also in Figure 4. Increasing X_F from 0.1 to 0.6 mol/mol, the number of stages required decreases slightly for FRT, differences between which, diminish at lower reflux ratio, R = 3. There was no difference in the number of stages for FRT when X_F was increased from 0.5 to 0.6 mol/mol for all values of reflux ratio. For

FBP, on the other hand, distinct curves were observed for all values of X_F from 0.1, 0.3,0.5 and 0.6 mol/mol. Moreover, at X_F= 0.6 mol/mol, a shift towards a lower number of stages was observed for FBP compared to FRT. Also, higher distillate purity was observed for FBP than FRT. Comparing the result for X_F = 0.6 mol/mol, for example, X_D = 0.85 was reached with N = 15, 12 and 10 for R = 3, 4 and 5, respectively, when FBF was used which could only be reached with N = 20 stages for the FRT. This observation shows that the FBT condition allows separation at low R and N, which is more favorable and efficient hence, less expensive compared to the FRT. Moreover, X_D values higher than 0.85 were reached for the FBP but not for the FRT for all values of reflux ratio. This was observed for X_F = 0.5 and 0.6 mol/mol and 0.3 mol/mol for R = 4 and 5, respectively. In general, the FRT was less sensitive to changes in X_F compared to FBT.

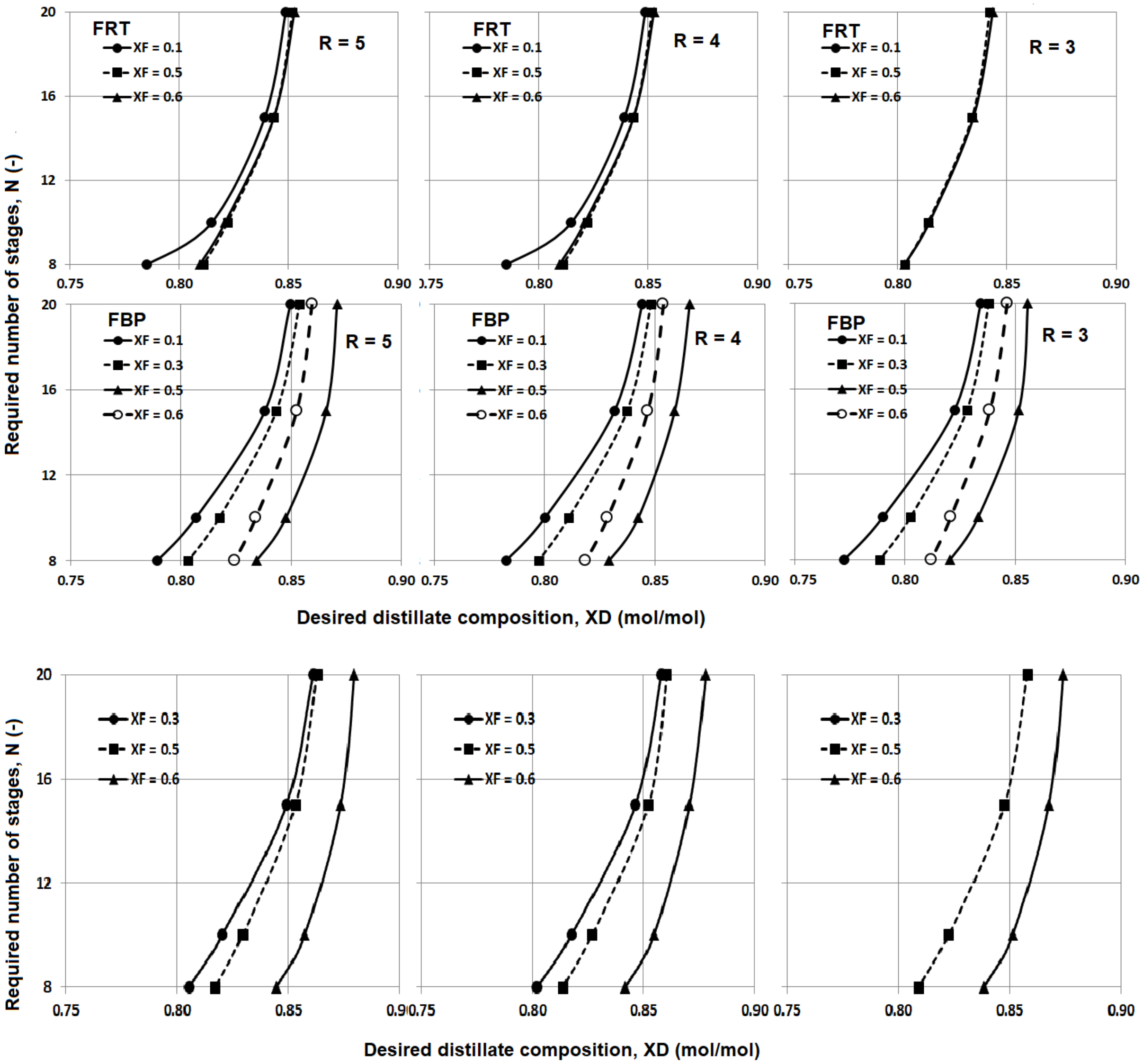


Figure 5: Effect of distillate purity on the required number of stages for the different FTCs (FRT, FBP and FDP) at various feed compositions, X_F, and reflux ratios, R.

Figure 5 shows further that the FDP leads to lower number of stages for higher X_D compared to FRT and FBP conditions. Distillate purity higher than 0.85 mol/mol was reached at a lower number of stages for all values of X_F = 0.3, 0.5 and 0.6 mol/mol. For example, with15 stages, X_D= 0.875 was reached at X_F = 0.6 mol/mol for all three levels of reflex ratio which was not observed for both FRT and FBP conditions. Hence, when aiming at higher distillate purity and keeping other variables fixed, the need for a large number of stages is attributable to the need for ethanol enrichment, which requires additional stages, because higher purity is achieved near the pinch point where VLE is closer to the $y = x$ line, leading to a more demanding separation, and large number of stages. Moreover, the driving force for mass transfer decreases towards zero when X_D = X_{AZ} (azeotropic point), that is, the concentration of ethanol in the vapor and liquid phases are nearly the same, necessitating a larger number of stages.

Figure 6 shows the effect of increasing distillate purity on the required number of stages at fixed reflux ratios and feed composition for different FTCs. The plots were made for distinct values of R= 4, 5 and 8 and various feed compositions. Results show that the reflux ratio strongly affects the number of stages by shifting the curves towards higher distillate purity for a similar range of the number of stages. This shows that higher distillate purity corresponds to fewer stages when the reflux ratio is increased from 4 to 8. By increasing the reflux ratio from 4, 5 to 8, the maximum distillate purity attainable increases from 0.847 to 0.862 mol/mol and finally to 0.880 mol/mol, respectively. For R = 8 and X_F = 0.5 mol/mol, the SHV leads to fewer stages compared to the FDP. This is because the SHV joins the vapor flow in the rectifying section directly without reaching the reboiler. Moreover, at R = 8 and X_F = 0.6 mol/mol, the FDP shows the least number of stages compared to the SHV. On the other hand, when R was set at 5 and X_F = 0.6 mol/mol, the number of stages decreased slightly from the FBP to the FRT.

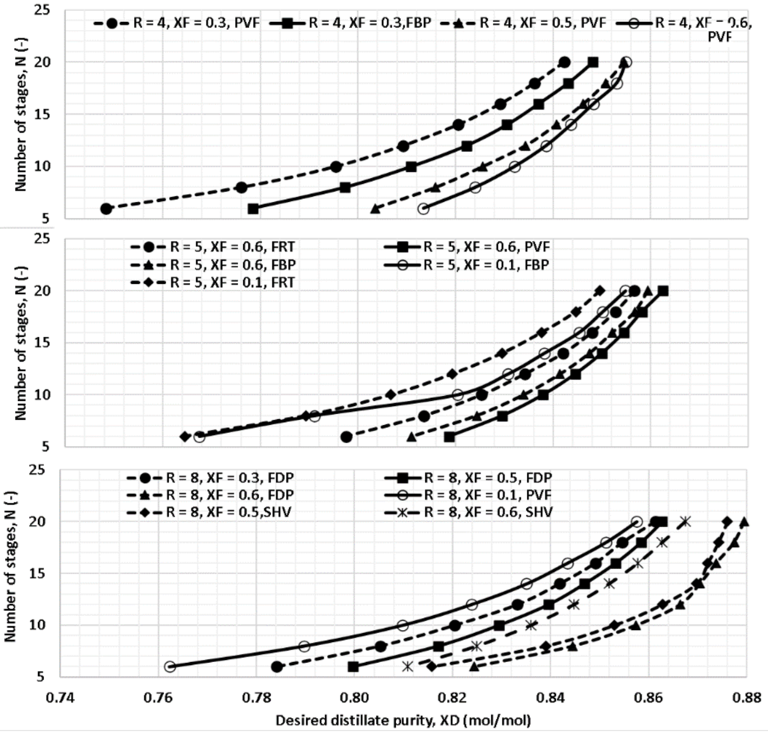


Figure 6: Variation of the number of stages with product purity, X_D, at different reflux ratios, feed compositions and FTCs.

The data parented in Figures 5 and 6 gives plant operators a tool for selecting the desired levels of operating conditions for high distillate purity during pre-concentration of the ethanol-water mixtures. For a given column, the number of stages is known, say 10, for which a proper choice of operating conditions can lead to high distillate purity. For instance, at R = 5 and X_F= 0.6 mol/mol, PVF leads to higher distillate purity, while at R = 8, the FDP leads to higher purity for the same feed composition.

4.2 Effect of distillate purity on reflux ratio

As stated earlier, the reflux ratio is the easily changed operating variable to attain high distillate purity, necessitating an insight understanding of the relationship between the two variables. This was studied by fixing the number of stages and feed composition. Figure 7 shows the effect of increasing distillate purity on the reflux ratio required to accomplish separation at a fixed number of stages (N = 8), four levels of feed composition and one selected FTC, that is PVF. The general feature of the plots is that the reflux ratio increases abruptly for a slight increase in distillate purity from R = 1 to 7 depending on the operating conditions. For FRT and PVF, the lowest reflux ratio required (R = 1) was observed for the highest concentration of solute in the feed, that is, X_F = 0.6 mol/mol. When the feed composition was decreased to 0.1 mol/mol, the curve shifted upward indicating that a higher reflux ratio was required to effect separation. Moreover, at the lowest feed composition, X_F = 0.1 mol/mol, the distillate purity achievable was observed to fall in a narrow range, from 0.77 to 0.80 mol/mol.

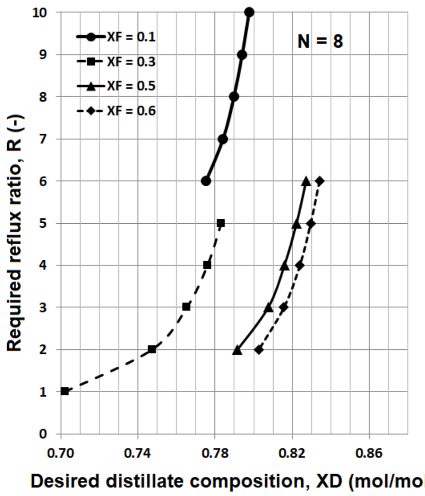
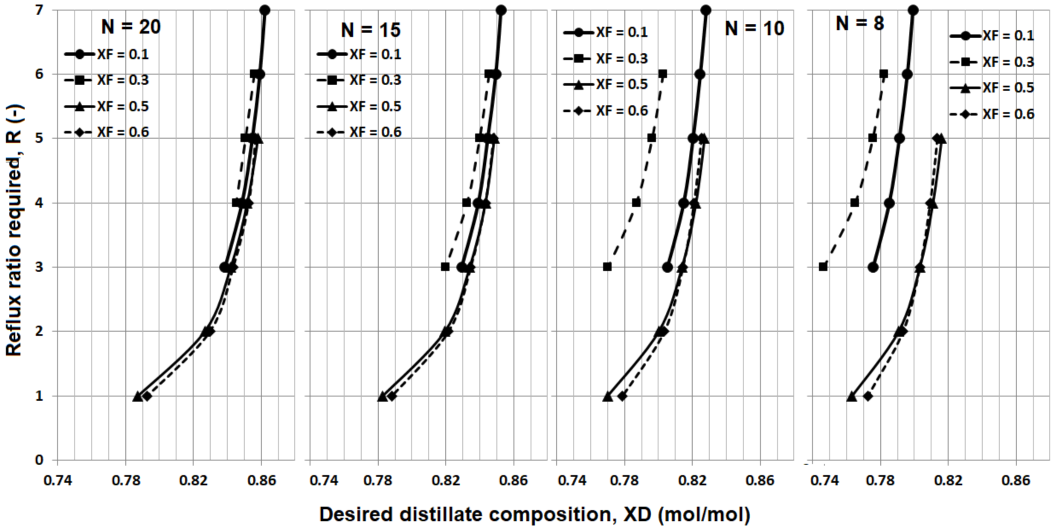


Figure 7: Variation of the required reflux ratio with the desired distillate purity for PVF (50% vaporized) for $N = 8$ stages.

Figure 8 shows the variation of reflux ratio with distillate purity for a fixed number of stages. Fixing the value of reflux ratio, that is moving horizontally, the highest distillate purity was reached at a higher concentration of the solute in the feed. The effect of feed composition was appreciable for10 and 8 stages for both FRT and PVF, different from a higher number of stages, that is, 20 and 15. With10 stages, for example, the effect of increasing X_F from 0.5 to 0.6 mol/mol did not show any appreciable difference. Figure 8, therefore, allows plant operators to select favorable values of R to maximize product purity without increasing the operation costs. For example, when feeding a 10-stage column with dilute feed (X_F= 0.1 mol/mol), the maximum distillate purity achievable was 0.80 mol/mol when the reflux ratio was set at 5. Therefore, to achieve higher separation efficiency with higher distillate purity at different FTCs, feed composition and number of stages, a higher reflux ratio is required to increase the contact intensity between vapors and liquid in the column (rectification). When a concentrated feed is used, a lower reflux ratio is required due to the high ethanol content in both vapor and liquid inside the column. Higher distillate purity also requires a higher reflux ratio to improve rectification but also to enhance the thermal efficiency of the distillation process. This aspect necessitates heat integration considerations to reduce utility consumption. When N is fixed, higher X_D implies operating the column towards a pinch point (closer to the azeotropic point) which requires a higher reflux ratio as the only controlling variable.

The dilute feed mixture ($X_F = 0.1$ mol/mol) required the highest reflux ratio (from $R = 6$ to 10) while for $X_F \geq 0.3$ mol/mol, the range of reflux ratio was lower (from 1 to 5 only) for the same range of distillate purity. Increasing the feed composition from 0.5 to 0.6 mol/mol, a horizontal shift was observed, that is, a higher distillate purity was achieved for a similar range of reflux ratio (2 to 6). Further analysis was conducted by varying operating conditions (N from 8 to 20 stages, X_F from 0.1 to 0.6 mol/mol, and FTCs covering FRT and PVF. It is interesting to note that a pre-concentration process can be initiated with a very dilute mixture, $X_F = 0.1$ mol/mol, leading to a high distillate purity, $X_D = 0.80$ mol/mol, by operating the column at higher reflux ratios ($R = 10$).



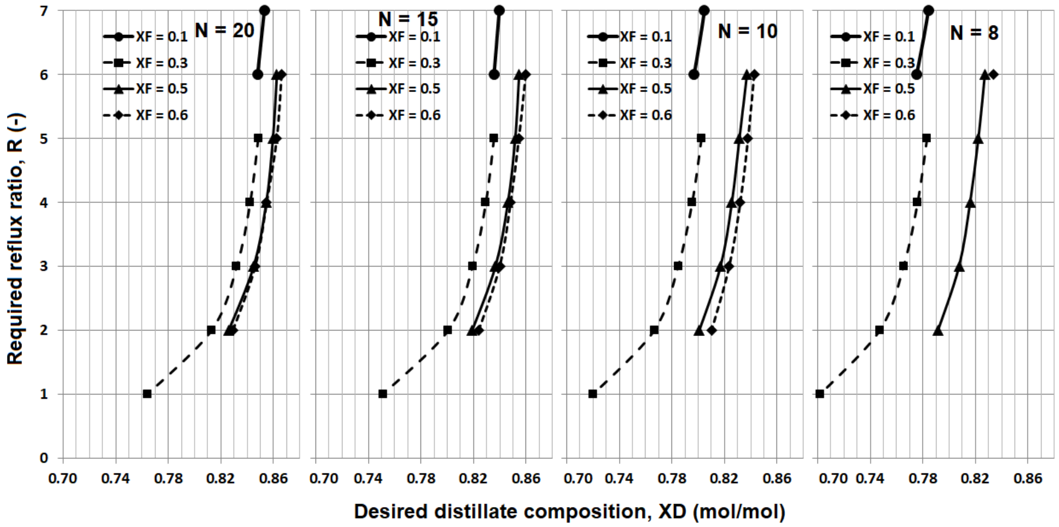


Figure 8: Variation of the reflux ratio required with the desired distillate purity for the FRT and PVF (50% vaporized).

4.3 Effect of increasing distillate purity on the condenser duty

Condenser duty signifies the required rate of heat removal from the column by cooling water to condense the vapors leaving at the top of the column. Once condenser duty, denoted as Q_c, is known and based on the heat transfer design equation for the condenser ($Q_c = UA\Delta T$) the water flow rate can be established based on inlet and outlet temperatures. Thus, a critical parameter for the design of the condenser is Q_c. This study presents the variation of condenser duty with distillate purity for a fixed number of stages, the FTCs and feed composition. Figure 9 presents the variation of condenser duty with different feed compositions. The data is plotted for PVF with 50% vaporization and for*8* stages. The condenser duty increases with X_D according to a 3rd order polynomial function for all operating conditions.

The condenser duty varied between 5 and 20 MW for F = 100 kmol/h, with X_F ranging between 0.1 and0.6 mol/mol. Further demand for distillate purity will also lead to higher condenser duty. Therefore, different operating conditions were studied to establish the settings leading to low Q_c but higher distillate purity as a performance target. The highest values for Q_c were observed at X_F = 0.3 mol/mol, and the lowest at X_F = 0.6 mol/mol, indicating non-uniformity in the effect of feed composition on condenser duty. A polynomial relationship between Q_c and X_D is an indication of a complex dynamic system, that can be described by using a non-linear relationship. This indicates further that the ethanol-water pre-concentration column is affected by a large number of variables, which is a good sign of a complex unit operation. The behavior shown by the process at X_F = 0.3 mol/mol requires further studies on the effect of X_F on the relationship between Q_c and distillate purity.

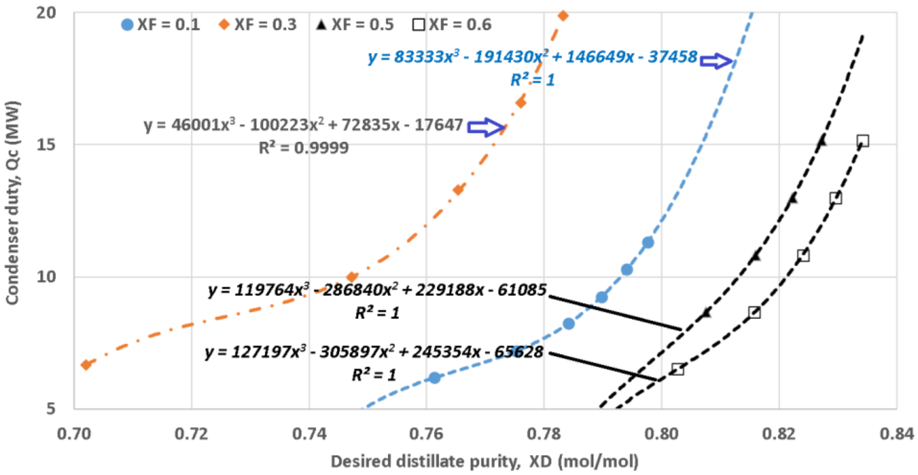


Figure9: Variation of the required condenser duty with the distillate purity for PVF (50% vaporized) for8 stages.

Figure 10 compares the values of $Q_c$at various stages for the FRT and PVF. Decreasing the number of stages from 20 to 8, the condenser duty increases, especially at X_F = 0.3 mol/mol when feed at room temperature was used. For the PVF, on the other hand, there were no appreciable changes in Q_c when the number of stages was decreased in the same range. Moreover, the highest values of Q_c were observed at X_F =0.3ranging from 5 to 20 MW. For FRT, however, the values of Q_c at X_F = 0.3 mol/mol were highest for the lower number of stages (N = 8 and 10). In general, the effect of X_F on Q_c is strong, a scenario that was observed for all operating conditions, as shown in Figure 10. The highest desired distillate purity X_D= 0.86 was observed for PFV at N = 20 and lower distillate purity at N = 8. For the largest number of stages (N = 20), similar X_D values were reached for both FRT and PVF. For PVF, however, higher distillate purity was reached at N =10 and 8 allowing for column operation at a lower number of stages. For columns with N = 10 say, PFV will be preferable to the FRT although the condenser duty will be higher than 5 MW.

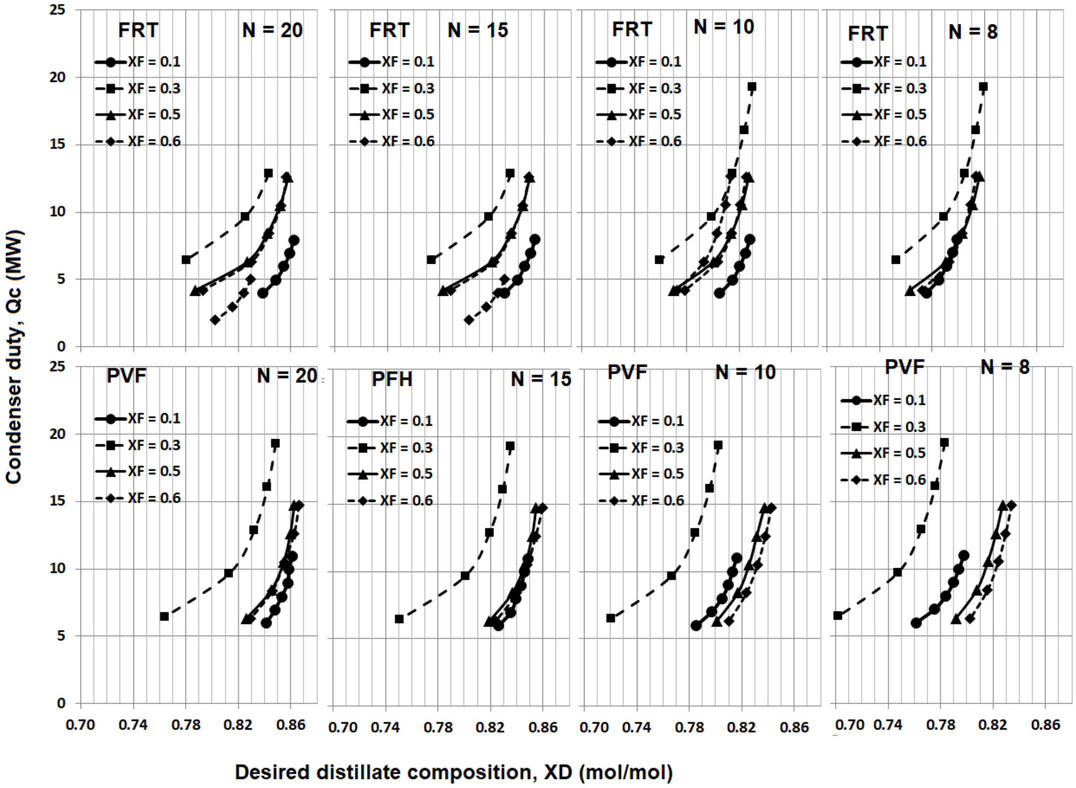


Figure 10: Variation of the condenser duty with the desired distillate purity for FRT and PVF thermal conditions.

Figure 11 compares the variation of condemner duty with X_D for FBP, FDP and SHV. Except for N = 20 stages where X_F = 0.3 mol/mol leads to the highest values of condenser duty, FBP leads to the lower condenser duties ranging between 2 and 15 MW only. For FDP and SHV the condenser duty values were higher, ranging between 12 and 25 MW, although higher distillate purity was attained. When the feed was at dew point or superheated vapor, the increase in condenser duty as distillate purity increased was the sharpest occurring in a narrow range of distillate purity values, especially when a column with a large number of stages is used. The FBP on the other hand, shows a wider range of distillate purity. For distillation columns with 20, 15 and 10 stages, FDP and SHV allow for higher distillate purity than FBP, which however operates at law condenser duties. Thus, the higher distillate purity attainable for the FDP and SHV is paid for by higher energy demand in the condenser, that is, increased heat load to be removed from the system. Results indicate that Q_c increases with increasing distillate purity for all fixed values of feed composition and reflux ratio. The need for high condenser duty in producing highly pure ethanol-water distillate is primarily due to the close boiling points, azeotropic behavior, hydrogen bonding, and the requirement for increased reflux to achieve the desired separation. These factors collectively and simultaneously contribute to the energy-intensive nature of achieving high-purity distillates in the system.

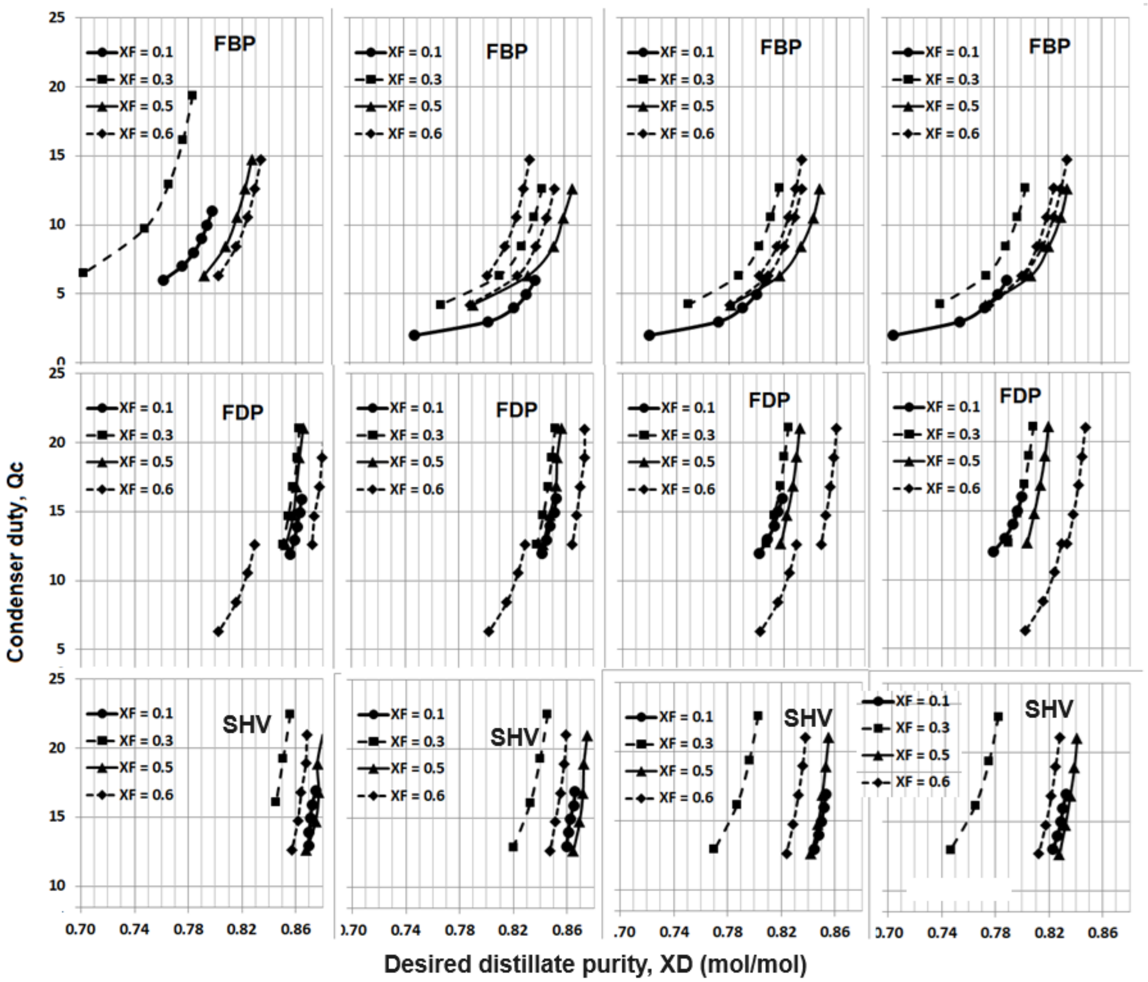


Figure 11: Variation of the condenser duty with the desired distillate purity for FBP, FDP and SHV feed thermal conditions.

There are several reasons for the sharp increase in condenser duty due to an increase in distillate purity for ethanol-water separation. Firstly, as the distillate purity increases, the relative volatility of the mixture decreases, and the *y*-*x* curve approaches the $y = x$ line. This leads to a higher boiling point of the mixture, which in turn requires a higher condenser duty to achieve the desired degree of separation. The required vapor load must also be increased necessitating higher reboiler temperature and reboiler duty to achieve higher vapor load to the condenser, which ultimately increases the condenser duty.

Secondly, the presence of the azeotropic point in the ethanol-water mixture also contributes to the sharp increase in condenser duty. As the distillate purity of the mixture increases, the azeotropic point moves closer to the desired ethanol concentration, which results in a uniform boiling/condensing mixture, whereby, both ethanol and water maintain constant presence in the vapor phase, increasing cooling load. Consequently, a higher condenser duty is required, which can significantly affect the energy consumption and operating costs of the separation process. Several studies have investigated this issue and proposed various methods to reduce the condenser duty and improve the energy efficiency of ethanol-water separation, such as energy efficiency improvements and use of deep eutectic solvents (Zhang *et al.*, 2019), use of solids of different design features as packings (Hassenrück *et al.*, 2017), energy-efficiency and heterogeneous distillation using macroporous resin (Li *et al.*, 2016), analysis energy consumption using Aspen Plus (Shukla *et al.*, 2019) and combination of both experimental and computer modeling to analyze effects of feed composition and packing heights (Moradi *et al.*, 2014).

4.4 Effect of distillate purity on reboiler duty

To produce distillate of high purity the energy requirement must be addressed especially in the reboiler, where steam is used to generate vapors from liquid mixtures. The supply of steam is rated by the power generated in MW which must be paid for. The energy requirements, however, depend on several variables for the distillation column, such as distillate purity, feed composition, the FTCs, reflux ratio and number of stages. Results show that the reboiler duty increased with increasing desired distillate purity for all fixed values of X_F, as shown in Figure 12. The data plotted in Figure 12 was collected for $N = 8$, when a PVF was introduced into the column. The reboiler duty increased with distillate purity according to 3rd order polynomial functions depending on the feed composition. Higher distillate purity and lower reboiler duties were observed for X_F= 0.5 mol/mol and lowest for X_F= 0.6 mol/mol.

Therefore, similar to condenser duty, the highest distillate purity and lowest reboiler duty were observed for a more concentrated feed. For PVF and *N* = 8 stages, the reboiler duty ranged between 1.0 and 15 MW. A third-order polynomial fit signifies a complex relationship between the two variables, influenced by the other variables as per the input/output block diagram shown in Figure 3.

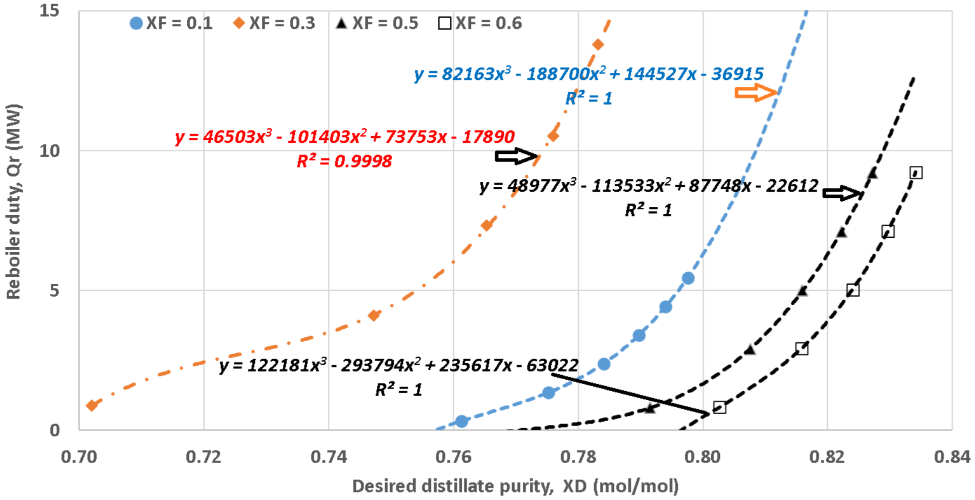


Figure 12: Variation of reboiler duty with distillate purity for PVF (50% vaporization) for *N* = 8 stages.

Figure 13 compares the reboiler duties at various distillate purities for different numbers of stages (N = 20, 15, 10 and 8 stages) and also for different FTCs (that is, FRT, FBP, SHV, FDP and PVR). The higher values of reboiler duty were observed at X_F= 0.3 mol/mol for all FTCs and the number of stages, while the lowest values were recorded at X_F = 0.6 mol/mol. For SHV, FDP and PVF, the increase in reboiler duty as the distillate purity increases was sharper compared to FRT and FBP, that is, for the former FTCs, the reboiler duty is more sensitive to *the* changes in distillate purity. When a feed at boiling point (FBP), for instance, was introduced into the column, the increase in reboiler duty was the slowest. Thus, SHV and FDP lead to very narrow ranges of X_D for the fixed values of *N* and X_F, especially for *N* = 20 and 15. The choice of operating condition which leads to higher distillate purity closer to 0.86 mol/mol can be identified in Figure 13 for *N* = 10 (for SHV and FDP), especially at X_F= 0.6 mol/mol. Using a very dilute mixture of ethanol and water (X_F = 0.1 mol/mol) the maximum distillate purity of 0.82 only could be reached. The sharp increase in reboiler duty due to the increase in distillate purity for ethanol-water separation is a critical observation. The reboiler duty is an important parameter in distillation operations, as it determines the amount of heat that needs to be supplied to the column to maintain the desired separation efficiency. As depicted in Figure 12, the profiles of reboiler duty versus distillate purity are nonlinear and are of similar shapes, as reported also by Qi *et al.* (2018). This observation requires detailed experimental and simulation studies.

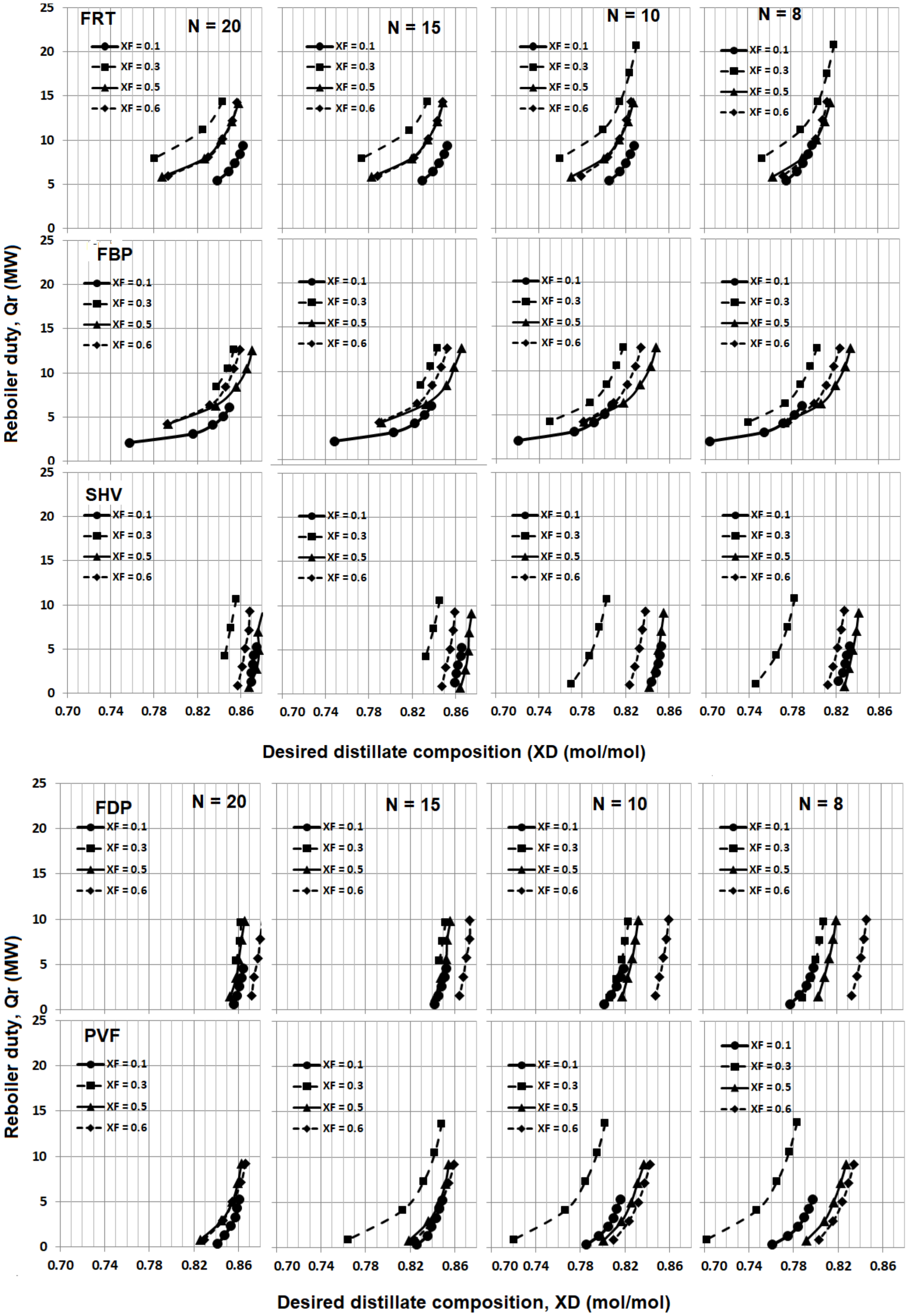


Figure 13: Variation of reboiler duty with distillate purity for different feed thermal conditions for *N* = 20, 15, 10 and 8 stages.

4.5 Effect of feed composition on reboiler and condenser duties

Figure 14 shows the effect of increasing feed composition X_F on both Q_c and Q_r for different FTCs and the number of stages. For Q_c, the effect of N was observed for FRT and SHV only at X_F= 0.3 mol/mol. Moreover, Q_r increased for X_F from 0.1 to 0.3 mol/mol, dropped and leveled again towards X_F= 0.5 and 0.6 mol/mol. For Q_r, a peak was also observed at X_F = 0.3 mol/mol for all FTCs, similar to Q_c. Moreover, the highest Q_r values were observed at X_F = 0.3 mol/mol for N = 15 (FDP and FBP). The variation of Q_r and Q_c with X_F shows a complex relationship when both N and FTCs were changed. For energy saving, the condition corresponding to X_F = 0.3 mol/mol should be avoided for all FTCs. There was no observed effect of N on Q_c for FDP, FBP and PVF. Moreover, the effect of X_F on both Q_c and Q_r was not appreciable for X_F = 0.5 and 0.6 mol/mol for all FTCs.

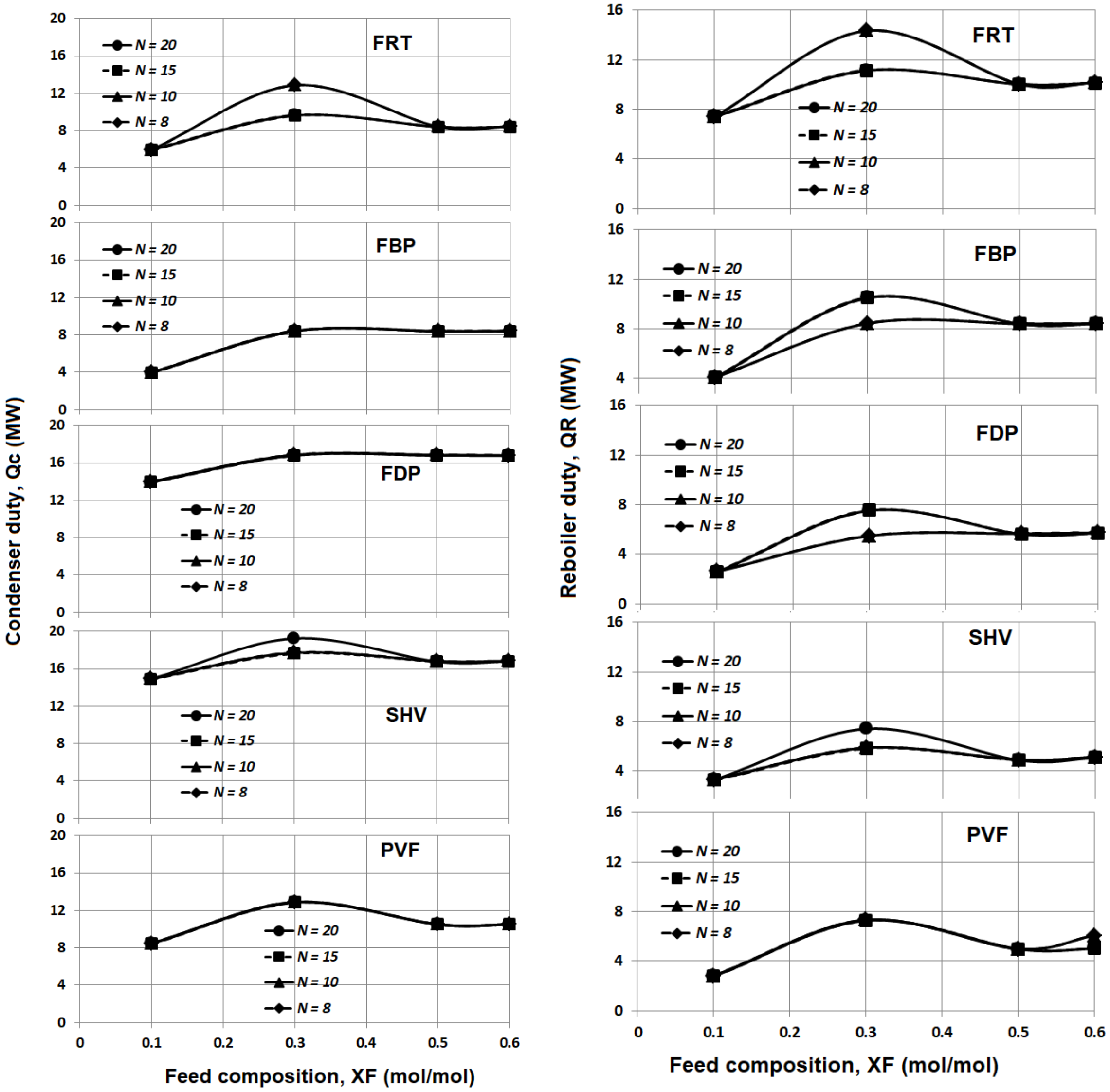


Figure14: Variation of the condenser and reboiler duties with feed composition during distillation of ethanol-water solutions before an azeotropic point for various feed thermal conditions.

The difference between energy input in the reboiler (Q_r) and the energy removed in the condenser Q_c was studied for different numbers of stages and feed composition. Figure 15 shows the values of (Q_r – Q_c) for different runs conducted at various feed compositions and the maximum and minimum number of stages. It was observed that the difference increases with X_F. That is, at higher feed ethanol concentration, Q_r was much higher than Q_c (at X_F = 0.6 mol/mol) than for dilute solutions, where the difference is smaller. This was observed for each fixed number of stages.

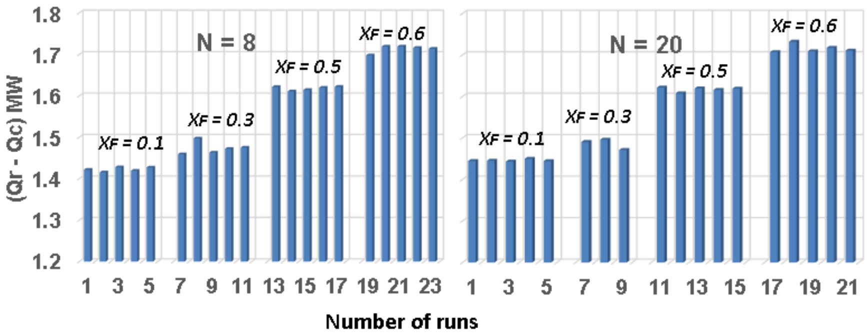


Figure 15: Variation of the difference between reboiler and condenser duties (Q_r - Q_c) with feed composition and the number of stages during pre-concentration of the ethanol-water mixture.

5. Conclusions

For all feed compositions, FTCs and reflux ratios, the number of stages increases faster with increasing distillate purity. For fixed values of X_F and N, the reflux ratio required to attain a given distillate purity increases with distillate purity. At higher feed composition, the reflux ratio required was lower compared to dilute feed solutions. For all operating conditions, the highest reflux ratios were required for very dilute feed composition (X_F = 0.1 mol/mol). The condenser duty increases with distillate purity according to 3rd order polynomial functions with good fit ($R^2 = 1$) for all fixed conditions of feed composition, number of stages and FTCs. Low condenser duty and high X_D values were observed when XF was highest for all operating conditions. For constant values of X_F, and number of stages, the condenser duty was highest for super-heated vapor (SHV) feed. The reboiler duty also increased with distillate purity according to the 3rd order polynomial function with good fit ($R^2 = 1$). The lowest values of reboiler duty were observed for SHV and FDP for all fixed number of stages and feed compositions. A comparison between the condenser duty and reboiler duty shows that the latter is higher for all operating conditions. The feed composition strongly affects the reboiler duty for a column with a lower number of stages (N = 8). The FDP and SHV show lower reboiler duty compared to other FTCs. The observation that slightly lower condenser and reboiler duties observed at $X_F \geq 0.5$ mol/mol compared to X_F = 0.1 mol/mol requires further investigation, as it shows a non-linear behavior. The difference between Q_r and Q_c (that is, Q_r - Q_c), increased with increasing feed composition for all fixed values of number stages, being highest at X_F = 0.6 mol/mol. Thus, even dilute solutions such as X_F = 0.1 mol/mol yield appreciable distillate purity when a normal distillation column is used. A very wide range of feed composition (0.1 to 0.6 mol/mol) has been investigated to show the effect of the desired distillate purity on the distillation process performance parameters. This study has covered a large number of variables affected by increasing the distillate purity, justifying the need for computer simulation in studying such complex non-linear systems.

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