STUDY OF THE THERMALLY COUPLED DISTILLATION SEQUENCES USING A NONEQUILIBRIUM STAGE MODEL

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A Petlyuk distillation column, considering equilibrium and nonequilibrium stage models, was studied. Rigorous simulations were conducted by using Aspen Plus^{TM} RATEFRAC Module for the separation of ternary mixtures. According to the equilibrium model, the energy-efficient design of the Petlyuk column requires that the intermediate component be extracted from the maximum point in the composition profile of the intermediate component in the main column. It was found that, for the intermediate component, mass transfer occurs from the vapour to the liquid phase from the top of the column to the stage where the sidestream is extracted, from this point mass transfer occurs in the opposite direction. This point, considering the nonequilibrium model, corresponds to the stage in which the net mass transfer rate is zero. For the case of two segments per stage, it was found that the heat duties predicted by the equilibrium model are significantly lower than those obtained by using the nonequilibrium model, which is consistent with previous reported results. However, it is important to say that despite of the higher energy duty predicted by the nonequilibrium model, both models predict significant energy savings.

KEYWORDS: thermally coupled distillation sequences, nonequilibrium model

INTRODUCTION

Distillation is the most widely used separation operation for most of the fluid mixtures. Unfortunately, not only distillation columns require a large amount of energy to achieve the separation task, but also this separation technique is highly inefficient in its use because of its low thermodynamic efficiency. As a result, the search for energy-efficient distillation schemes is a current trend in process systems engineering. One way of reducing the energy demand in distillation is by using thermally coupled distillation sequences. An important effort has been focused on the development of new design and optimization methods for thermally coupled distillation columns, which can provide savings up to 30% of the total annual cost for the separation of some multicomponent mixtures, as compared to classical distillation sequences based on conventional columns (Tedder and Rudd, 1978; Hernández and Jiménez, 1999; Triantafyllou and Smith, 1992, among others). Such coupled distillation sequences use thermal links that can be implemented by eliminating either a reboiler or a condenser and then introducing recycle streams in the vapour or the liquid phases, respectively. The most important thermally coupled distillation sequence is the Petlyuk column (fully thermally coupled) which

uses a prefractionator linked by two recycle streams (Figure 1). This distillation sequence has been implemented in some chemical industries through the use of a single shell and a dividing wall, and the reported savings in both energy and capital costs have been of around 30% (Petlyuk et al., 1965). Its thermodynamic efficiency has been attributed to the absence of remixing in the main column, i.e., the sidestream is placed where the maximum concentration value of the composition profile of the intermediate component is located (Hernández and Jiménez, 1999). Design methods for the Petlyuk column must take this fact into account in order to guarantee the energy consumption reduction. Most of the works related to the design, optimization and control of distillation columns use the equilibrium model approach, obtaining, for the case of the Petlyuk column, energy savings of up to 50%. However, no use of the nonequilibrium model approach has been made so far for the study of the Petlyuk column. As a result, the simulation of this column by using a rigorous nonequilibrium stage model is carried out in this work in order to understand its steady state behavior.

The basic equations for the nonequilibrium model can be found elsewhere (e.g. Seader and Henley, 1998). These equations include phase mass balances, phase energy balances, equilibrium relations, summation equations, mass transfer rate in the vapor phase, mass transfer rate in the liquid phase, and energy transfer rate. It is worth noting that some works regarding nonequilibrium stage model have been reported, but these works only deal with single columns. Important works were published by Krishnamurthy and Taylor (1985a; 1985b). They used the nonequilibrium stage model in the simulation of distillation columns using some solution techniques based on Newton's method. They also
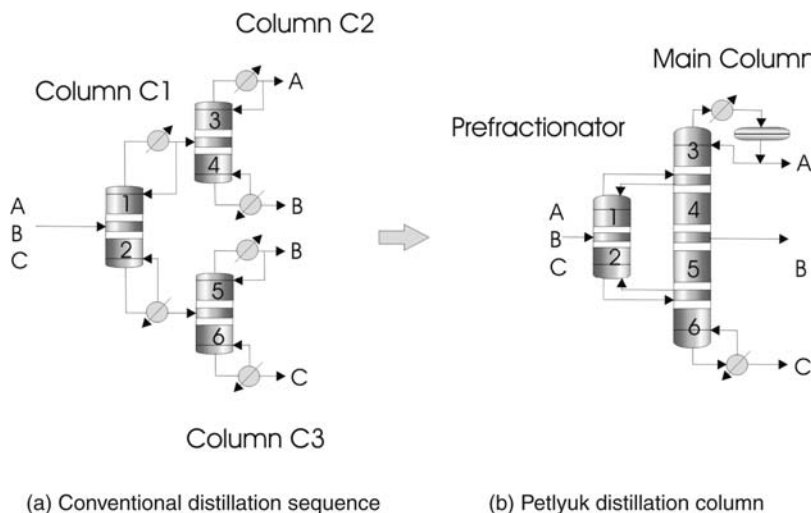

(a) Conventional distillation sequence (b) Petlyuk distillation column

Figure 1. Design strategy of the Petlyuk column from a conventional distillation sequence

compared their results with experimental data. Taylor *et al.* (2003) extended the application of the nonequilibrium approach for the modeling of different distillation cases, and they found that this representation can convey more realistic results, even for complex distillation, e.g. reactive distillation, three-phase distillation, heterogeneous azeotropic distillation, etc. Higler *et al.* (2004) applied the nonequilibrium model to the three-phase distillation case, including mass and energy balances for each of the three phases. They found that a nonequilibrium model is more suitable for this class of problems than the equilibrium model, even considering efficiency factors. A nonequilibrium model for three phase distillation in packed distillation column was presented by Repke *et al.* (2004). Their model also took into account the mass transfer between all phases.

In this work, the application of the nonequilibrium stage model to the simulation of a Petlyuk distillation column is presented. The Aspen Plus^{TM} RATEFRAC Module was used to obtain the composition profiles, mass transfer rates and optimum energy duty for this complex column.

NONEQUILIBRIUM MODEL

In the nonequilibrium model, the mass and energy balances around each stage (Figure 2) are replaced by separate balances for each phase around the stage, which can be a tray, a collection of trays, or a segment of a packed section. In residual form, the equations are as follow:
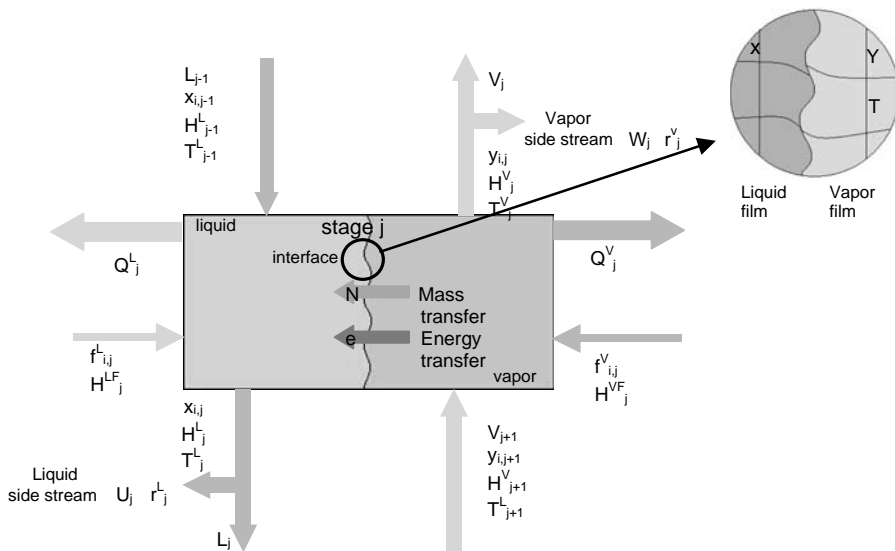

Figure 2. Nonequilibrium stage

Liquid-phase component material balances:

$$M_{i,j}^L \equiv (1 + r_j^L)L_j x_{i,j} - L_{j-1} x_{i,j-1} - f_{i,j}^L - N_{i,j}^L = 0, \quad i = 1, 2, \ldots, C \qquad (2.1)$$

Vapor-phase component material balances:

$$M_{i,j}^V \equiv (1 + r_j^V)V_j y_{i,j} - V_{j+1} y_{i,j+1} - f_{i,j}^V + N_{i,j}^V = 0, \quad i = 1, 2, \ldots, C \qquad (2.2)$$

Liquid-phase energy balance:

$$E_j^L \equiv (1 + r_j^L)L_j H_j^L - L_{j-1} H_{j-1}^L - \left(\sum_{i=1}^C f_{i,j}^L \right) H_j^{LF} + Q_j^L - e_j^L = 0 \qquad (2.3)$$

Vapor-phase energy balance:

$$E_j^V \equiv (1 + r_j^V)V_j H_j^V - V_{j+1} H_{j+1}^V - \left(\sum_{i=1}^C f_{i,j}^V \right) H_j^{VF} + Q_j^V + e_j^V = 0 \qquad (2.4)$$

For the vapor-liquid interface, I

$$E_j^I = e_j^V - e_j^L = 0 \qquad (2.5)$$

Equations (2.4) and (2.5) are coupled by the component mass-transfer rates:

$$R_{i,j}^L = N_{i,j} - N_{i,j}^L = 0 \quad i = 1, 2, \ldots, C - 1 \qquad (2.6)$$

$$R_{i,j}^V = N_{i,j} - N_{i,j}^V = 0 \quad i = 1, 2, \ldots, C - 1 \qquad (2.7)$$

The equations for the mole fraction summation for each phase are applied at the vapor-liquid interface:

$$S_j^{LI} = \sum_{i=1}^C x_{i,j}^I - 1 = 0 \qquad (2.8)$$

$$S_j^{VI} = \sum_{i=1}^C y_{i,j}^I - 1 = 0 \qquad (2.9)$$

Phase equilibrium for each component is assumed to exist only at the vapor-liquid interface:

$$Q_{i,j}^I \equiv K_{i,j} x_{i,j}^I - y_{i,j}^I = 0 \quad i = 1, 2, \ldots, C - 1 \qquad (2.10)$$

Because only C-1 equations are written for the component mass-transfer rates in (2.6) and (2.7), total material balances in terms of total mass-transfer rates, $N_{T,j}$ can be added to the system:

$$M_{T,j}^L \equiv (1 + r_j^L)L_j - L_{j-1} - \sum_{i=1}^{C} f_{i,j}^L - N_{T,j} = 0 \qquad (2.11)$$

$$M_{T,j}^V \equiv (1 + r_j^V)V_j - V_{j+1} - \sum_{i=1}^{C} f_{i,j}^V + N_{T,j} = 0 \qquad (2.12)$$

$$N_{T,j} = \sum_{i=1}^{C} N_{i,j} \qquad (A.13)$$

DESIGN AND OPTIMIZATION METHODS

The energy-efficient design of the Petlyuk column was obtained using the equilibrium-based method described by Hernández and Jiménez (1999). The method is briefly depicted in Figure 1. The conventional distillation sequence shown in Figure 1a results from the use of the Fenske-Underwood-Gilliland shortcut distillation method, optimized by using the rigorous equilibrium stage model included in the Aspen Plus[TM] RADFRAC Module. The number of prefractionator stages shown in Figure 1b are equal to the number of stages in column C1 from Figure 1a (sections 1 and 2). The total number of stages in the main column of the Petlyuk sequence (Figure 1b) equals the summation of the stages in columns C2 (sections 3 and 4) and C3 (sections 5 and 6) shown in Figure 1a. Two recycle interconnecting streams are introduced in the Petlyuk sequence. The mass flowrate of both recycle streams is varied until the minimum energy requirement in the reboiler is found. This optimization strategy is explained in detail in the work of Hernández and Jiménez (1999).

CASE OF STUDY

To assess the application of the nonequilibrium stage model, the separation of a ternary mixture of n-butane, n-pentane and n-hexane with molar fractions of 0.4, 0.2 and 0.4, respectively, is considered. Recoveries of up to 98.7%, 98% and 94%, respectively, were obtained for each component of the ternary mixture. Three different types of distillation trays were tested: sieve, valve and bubble cap. Operational pressures were set in advanced in order to guarantee the use of cooling water in the condensers.

RESULTS

The study was conducted in two stages: in the first part, the design and optimization of the Petlyuk column were obtained by using a rigorous equilibrium stage model, whereas in the second part, the optimized design through the use of the nonequilibrium stage model was tested. The tray sections were determined through the use of the design and optimization

method reported by Hernández and Jiménez (1999) assuming equilibrium operation. When the nonequilibrium stage model was used for the simulation of the Petlyuk column, an important aspect to note is that the sidestream is obtained from maximum point in the concentration profile of the intermediate component. This guarantees a good design with respect to the energy consumption (although it may not be the global optimum value). Figure 3 shows the minimization of the energy consumption for the two recycle streams by using the nonequilibrium model. It is important to note that the optimization surface is very similar to that obtained using the equilibrium model, as reported in the work of Jiménez *et al.* (2001). The optimization was carried out considering two segments per each stage, this is in agreement with the work of Peng *et al.* (2002). For instance, in Figure 4 it can be observed that mass transfer occurs from the vapour to the liquid phase as we move from the top to the sidestream tray and in the opposite direction from this to the bottom tray. In stage 17, the net mass transfer rate is zero, which corresponds to the maximum point in the composition profile. The net mass transfer rate for the lightest component occurs from the liquid to the vapor phase in the upper part of the column and it is zero in the lower part. For the case of the heaviest component, the net mass transfer is zero from the top to sidestream stage, and it occurs from the vapour to the liquid phase in the rest of the column. Since the effective mass transfer area is very important in the case of the nonequilibrium stage model, the influence of the column diameter on the energy consumption is shown in Figure 5. This behavior is not predicted by the equilibrium stage model. It can be observed that the energy consumption is a strong function of the type of plate, being the lowest for the valve plate. Another
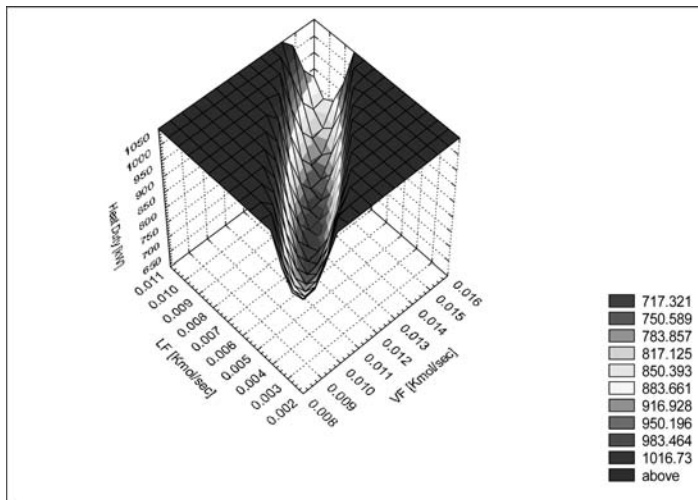

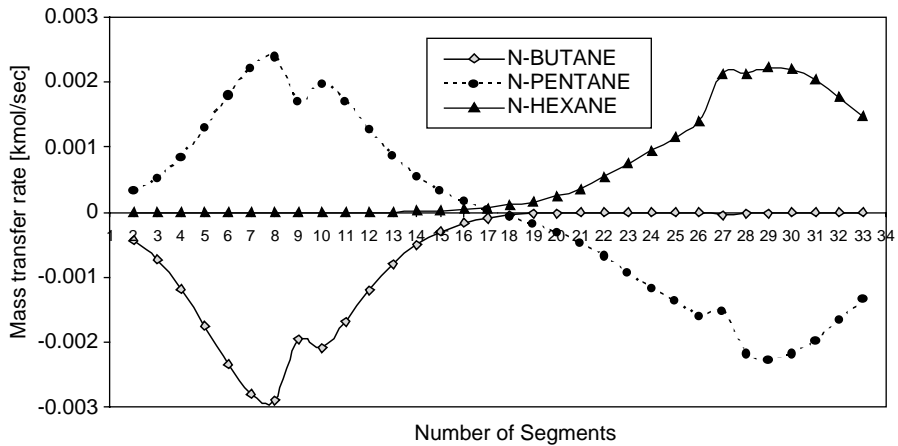**Figure 3.** Optimization of the Petlyuk column (nonequilibrium)

Figure 4. Mass transfer rates in the Petlyuk column (nonequilibrium)

important effect to be considered is the energy-performance of the Petlyuk distillation column and the conventional distillation sequence. In this sense, the equilibrium stage model predicts energy savings of up to 50% more for the Petlyuk distillation column than for the conventional sequence. It is important to say that the same number of stages and segments were considered in both cases. The results indicate that savings of
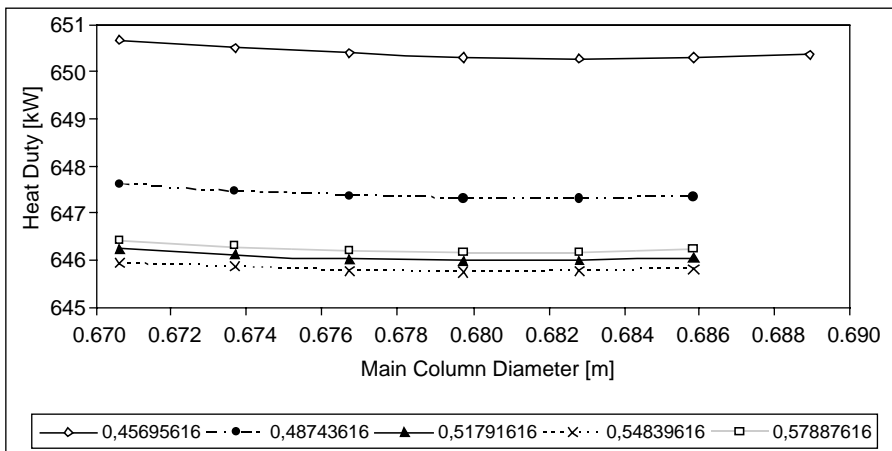

Figure 5. Influence of the column diameter in the energy consumption of the Petlyuk column

around 50 and 40% for the Petlyuk column were predicted by using the equilibrium and nonequilibrium models, respectively. The heat duties are significantly lower when the distillation sequences are modeled by considering the equilibrium model; however the values obtained by using the nonequilibrium model are more realistic.

CONCLUSIONS

In this work, a simulation of a Petlyuk distillation column and a conventional sequence for the separation of a ternary mixture of n-butane, n-pentane and n-hexane is presented. The simulation was carried out using both the equilibrium and nonequilibrium stage models for sieve, valve and bubble cap trays. The results indicate that both stage models predict significant energy savings and that the energy consumption depends strongly on the interconnection recycle streams. However, the equilibrium model predicts heat consumptions significantly lower than those obtained by assuming nonequilibrium operation, being the later more realistic. The dependence of the energy consumption on the diameter of the distillation column can only be predicted by the nonequilibrium stage model. It is important to note that the sidestream in the main column for the Petlyuk sequence should be placed where the maximum point in the composition profile for the intermediate component is located. In the case of the nonequilibrium stage model, this point corresponds to the zero net mass transfer rate.

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