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Distillation top-connected with ultrasound for separation of Ethanol/Water mixture

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Abstract: Recent and ongoing research in the distillation of non-ideal mixtures is reviewed focusing on advances in the methodologies for the synthesis, design, analysis and control of separation sequences involving homogeneous and heterogeneous azeotropic towers. This paper presents special combination processes of a distillation column and ultrasound flash distillation (USF) were consider. Simulation design is built on Aspen Plus as user defined unit operations for these two types of modules. The USF model was prepared to break the azeotrope of minimum boiling azeotropes and also the limitation of this model was discussed. Top stream of a distillation column is fed to the number of USF modules which connected serially with intermediate condenser were used to achieve purity with 99 mol% ethanol for separation ethanol/water mixture. The utilities of the distillation column and USF module such as cooling water, steam and electricity are selected to minimize the operating cost of the ultrasound assisted distillation process.

Keywords: ultrasonic waves, distillation column. azeotropic mixture, Optimization, aspen plus.

Introduction

The separation of azeotropic mixtures is an inherent part of biochemical and chemical processes and still remains a challenging issue in terms of process design and identification of selective separations. Well-known separation processes such as extractive, azeotropic distillation and pressure swing distillation often require high investment and energy costs. Membrane processes such as pervaporation and vapor permeation can be regarded as an alternative to established separation processes due to their high selectivity and their potential to achieve significant energy savings. However, membrane processes are not often used as stand-alone processes on the industrial scale because of high costs or low throughput. Systematic separation process design has mainly focused on single units operation, mostly distillation owing to its paramount importance in the process industries. If a separation is difficult or even impossible using a single type of unit operation, different separation principles can be combined to render the separation feasible or to improve its performance. Distillation is not particularly attractive for “difficult” separations involving mixtures of components with close boiling points or mixtures forming azeotropes [1, 2]. However, distillation has been used extensively even in these cases and significant academic and industrial research effort has been directed toward the development of efficient distillation-based systems for difficult separations.

A popular approach is to take advantage of multiple processing technologies and configure a hybrid process with multiple technologies so that the limiting behavior is overcome. A hybrid separation process is defined as the combination of at least two different unit operations in different apparatuses, which contribute to the separation task to achieve a predefined task [3, 4]. Some of the more common hybrid distillation processes include the distillation-adsorption [5-7], distillation-crystallization [8-10] and membrane distillation [11-13] processes. New approach is to intensify process by adding ultrasonic equipment to the separation process [14-18]. This technology has received an increasing attention in chemical process industry because cavitation as a source of energy input for chemical processes is increasingly being studied owing to its ability to generate rapidly formed and disappearing hot-spots under nearly ambient conditions. Thereby this technique offers significant energy savings along with substantial capital and space reduction. Since distillation is the basic separation system, interlinked a distillation column with ultrasound equipment to achieve a better, cheaper, easier and enhanced separation of azeotropic mixtures. When the applying ultrasonic waves, the positively change of vapor-liquid equilibrium and alter the properties of azeotropic mixtures [19, 20].

The main objective of this work is to develop a mathematical model of a single stage vapor-liquid equilibrium system in Aspen Custom Modeler is exported to Aspen Plus to represent one stage of ultrasound flash distillation (USF). The top stream of a distillation column is fed to the number of USF modules which connected serially with intermediate condenser in order to configure ultrasound assisted distillation process for separating ethanol-water mixture.

Design Methods and Tools

Aspen Plus^{TM} is the software package, frequently used in industry and academe [21]. This is a steady-state sequential modular simulation package used as a tool to simulate and design chemical processes [22]. It offers the possibility to simulate various combinations of unit operations such as reactors, distillation towers, heat exchangers and compressors using the built-in process models. Although a built-in stand-alone model for ultrasound equipment is not available in the standard version of Aspen Plus, this makes the user completely dependent on the way Aspen Plus handles this model and makes it difficult to direct the simulation in response to the results. With the help of the programming language Aspen Custom Modeler (ACM), one can build a program of sonication model and export it to a flowsheet in Aspen Plus, containing a different separation units a combined processes can be simulated.

The ability of sonication phenomena in facilitating the separation of azeotropic mixture presents opportunities for developing distillation system that is more intensified and efficient. It has already been experimentally illustrated in previous work [23] that the ultrasonic waves can significantly enhance the vapor liquid equilibrium (VLE) data of ETOH/ ETAC mixture by shifted of azeotropic point upwards from the standard curve and enhance the relative volatility leads to the separation of this mixture at high purity. To expedite further the much-needed developments, a mathematical model [24] of the system based on conservation principles, VLE and sonochemistry was developed. The model that was founded on a single stage VLE system and enhanced with ultrasonic waves was validated with experimental data. The model was coded by using MATLAB simulation tool and sensitivity analyses were carried out on ETOH/ ETAC mixture. Outcomes from model study concurred with experimental results and were consistent with theory. It can be concluded that the experimental and modeling work have successfully show that the ultrasonic wave has potential to break the azeotrope. A lower frequency and a higher intensity are preferred by choosing the right operating condition to separate azeotropic mixtures in a single column. These studies demonstrates that azeotropic point of this mixture was totally eliminate at the optimum values of sonication operating conditions including intensity of 500 w/cm^2 and frequency of 25 kHz.

Ultrasonic Assisted Distillation Process

A. Design Concept

Pressure has a marked effect on the VLE diagrams of azeotropic mixtures. Several literatures [25-28] show the VLE data of azeotropic mixtures at low pressure exceeded the consistency criteria, due to the association effects were not considered when correlating the VLE data. The model has been derived based on these principles taking into consideration of conservation thermodynamic equilibrium and sonication phenomena using the VLE studies previously carried out [23, 24]. Aspen Plus offers the possibility to simulate configuration design that is combination of different unit operations. Figure 1 shows that the USF modules are located to treat the product stream and separation is carried out in the distillation column. A single conventional distillation column with number of sonication modules (USF) arranged in series with intermediate condenser, receiving feeds from top of a distillation column and sending the outputs to the number of ultrasound series units. The feed enters the first stage of the USF, and the vapor stream produced from each stage condenses to become the feed to the stage above it; while the liquid bottom stream of each stage refluxes back to the stage before it.

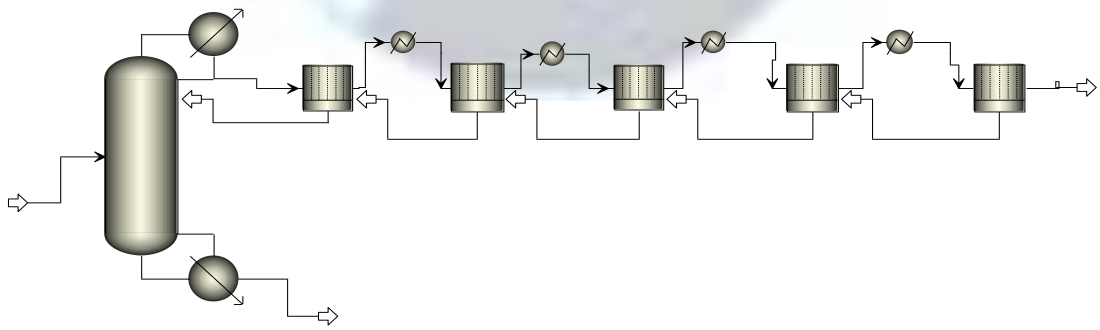

Figure 1. Schematic flow-sheet of the configuration represent series arrangement of ultrasound modules connected with top stream of a distillation column.

B. Simulation Study

In this study, the classic example of minimum boiling azeotrope is the ethanol and water mixture was selected to represent a case study for separated by proposed configuration. This mixture is known to form azeotrope of 89.4 mol% ethanol that boils at 78.2 ºC. The NRTL model was adopted for the calculation of the activity coefficients of the liquid

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phase, whereas ideal behavior was assumed for the vapor phase [29]. It is assumed that 65 mol% ethanol in feed composition at saturation stream with fixed in flow rate of 100 Kmol/h and at pressure 1 atm was chosen randomly.
The feed stream of the mixture enters into a normal distillation column and flow-sheet of simulation for azeotropes separation was illustrated in Figure 1. In order to obtain a distillate with close to azeotropic point from distillation column, the fixed specifications for the distillation column are considered 40 theoretical trays and the feed placed in tray number 31 (starting from the top of the column) with reflux ratio 5.

In order to shift the azeotrope, the top product of distillation column just sends it to few numbers of USF modules to obtain higher purity product. Figure 2 shows the azeotrope of ethanol/water mixture shifted upward and increased nonlinearly with increase of number of USD. It can be noted that a higher ethanol purity of 99 mole % were obtained with just 11 stages of USF.

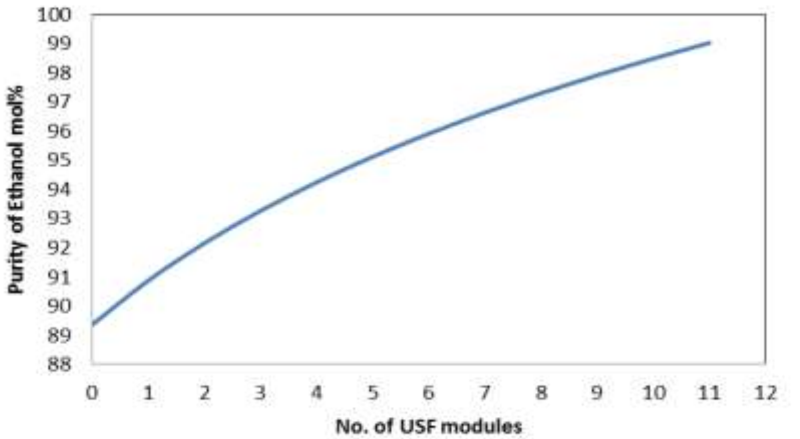

Figure 2. Purity of ethanol production related with number ultrasound units after distillation column.

Process Limitation

Generally, the basic principle for separation any type of azeotrope depends on three concepts: pressure variation such as pressure swing distillation; add third components as separating agent such as extractive distillation; and molecules size of a mixture such as membrane technique. The separation of minimum boiling azeotrope by a distillation column will result in the distillate being closer in composition to the azeotrope than the starting mixture. For example, when distilling a mixture of ethanol and water that is richer in ethanol than the azeotrope, the distillate will be poorer in ethanol than the original this means the bottom product will be richer in ethanol. Unlikely, in case of a maximum boiling azeotrope, when a light component in the feed mixture is richer than the azeotrope, it can be getting a still richer vapor and a fractionating column can get pure product out of the top [30]. Theoretically, In order to break the azeotrope of maximum boiling azeotropic mixtures by pressure variation, high pressure required to achieve the shifting of azeotropic point downward [31-34]. However, sonication technique is a model- basic concept of cavitation bubbles which are present micro-point of low pressure in liquid medium [35-37]. Therefore, the major limitation of the ultrasonic assisted distillation process is that in the separation of maximum boiling azeotropes, when a light component in the feed mixture is less than the azeotrope.

Another overcome of the proposed design is to separate a mixture that has feed composition close to azeotrope. Changes in the feed composition represent the most significant upsets with which a distillation control system must deal on a continuous basis. A feed composition change shifts the composition profile through the column resulting in a large upset in the product compositions. Many literatures and books refer to the separation of an azeotropic mixture by a normal distillation column will result in the distillate being close to the azeotrope. Therefore, if feed compositions are close to the azeotrope, ultrasonic assisted distillation process would not be possible because it is uneconomical.

Optimization

The objective of the optimization was to determine the economical process of ultrasonic assisted distillation design for the given separation task. To achieve this aim, an objective function that includes investment and operating costs is necessary. The investment cost of the proposed design involves the cost of equipment for the distillation column and ultrasounds modules. In case of distillation column the equipment cost including the cost of trays, tower, heat exchangers (reboiler and condenser) and reflux drum. In case of sonication unit the equipment cost including the cost of flash drum module, transducer, and intermediate condensers. Due to the cost of the equipment are not available and also cost of the raw material is fixed, in this work the operating cost of utility only was considered that involves the cost of cooling water, steam, and electricity.

The operating cost (C_o) of the ultrasonic assisted distillation process is calculated as the summation of column (C_{column}) and sonication modules ($C_{ultrasound}$) costs as follow:

$$C_o = C_{column} + C_{ultrasound} = (C_c + C_r + C_u) \quad [\$/h] \qquad 1$$
$$C_c = W_c * Q_c \qquad 2$$
$$C_r = S_r * Q_r \qquad 3$$
$$C_u = \sum_1^n E_u * Q_u + \sum_1^n W_c * Q_c \quad [\$/h] \qquad 4$$

Where Q_c (kJ/h) is the heat withdrew from the condenser of the distillation column and also of the ultrasound unit, Q_r (kJ/h) is the heat added to the reboiler of the column, Q_u (kJ/h) is the heat power generated by ultrasound, W_c ($/ton) is the cost coefficients of cooling water used in a condenser for the distillation column and to condensate the vapor between ultrasound units, S_r ($/MMBtu) is the cost coefficients of steam used in a reboiler for the column, E_u ($/kWh) is the cost of electric consumption of the ultrasound unit and n is the number of sonication modules used.
The operating cost of the processes is calculated by using equations 1 to 4 and the cost coefficients can be found in Seider et al [38]. From this book the price of cooling water which used to condense the distillate of the column and between the sonication modules is 0.0251 $/Ton, the price of low pressure steam (6 bar and 150 ^oC) which used to heat the bottom of the column is 5 $/MMBtu and electric supply to the ultrasound equipment is 0.05 $/kWh. A detailed summary of the results for operation cost of proposed configurations processes can be calculated by Aspen Plus as reported in Tables 1.

Table 1: operation cost of the configurations of ultrasonic assisted distillation processes

Item	Operation cost ($/h)	Percentage %
Distillation Condenser	7.2929	4.54
Distillation Reboiler	88.351	55.06
Cost for distillation column	95.6439	59.6
intermediate condenser of 11 modules	0.412	0.256
Electricity of 11 modules	64.46	40.15
Cost for sonication modules	64.872	40.4
Total	160.515	100

Conclusion

This work has successfully shows that the ultrasonic wave has potential to enhance the separation process of azeotropic mixtures with higher purity product. In the present study, created ultrasound flash distillation model (USF) in Aspen Custom Modeler and the model have been exported to Aspen Plus as a user defined unit operation. In order to separate ethanol/water mixture with purity of 99 mol% ethanol, number of the USF modules connected in series with intermediate condenser was required which are connected with the top product of a distillation column to obtain ultrasound assisted distillation configuration. However, this design has been unsuccessful to separate the maximum boiling azeotropes. Another overcome of the proposed design is uneconomical to separate a mixture that has feed composition close to azeotrope. These findings set a good beginning towards the development of an ultrasonic potential that is currently in progress.

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