Simulation and Optimization of Ethyl Acetate Reactive Packed Distillation Process Using Aspen Hysys

Abdulwahab GIWA and Süleyman KARACAN

Ankara University, Faculty of Engineering, Department of Chemical Engineering, 06100, Ankara, TURKEY
E-mail: agiwa@eng.ankara.edu.tr, karacan@eng.ankara.edu.tr

Abstract: The simulations and optimizations of a reactive packed distillation process for the production of ethyl acetate, water being the by-product, from the esterification reaction between acetic acid and ethanol has been carried out in this work using Aspen HYSYS 3.2 process simulator. The main column, apart from the condenser and the reboiler, was divided into five sections: rectification, acetic acid feed, reaction, ethanol feed and stripping sections. In the simulations, Non-Random Two-Liquid model was used as the fluid package and the reaction occurring in the reaction section of the column was modeled as an equilibrium one. In order to validate the results of the simulator, experiments were carried out in a reactive packed distillation pilot plant. The data input and output of the experiments were done with the aid of MATLAB/Simulink via electronic modules. The results of Aspen HYSYS simulations were compared with those obtained from the experimental studies.

Key words: Reactive packed distillation; Esterification reaction; Aspen HYSYS; Simulation; MATLAB/Simulink.

Introduction

In recent years, integrated reactive separation processes have attracted considerable attentions in academic research and industrial applications, (Völker, Sonntag, and Engell 2007). One of these processes which is known as "reactive distillation" is potentially attractive whenever conversion is limited by reaction equilibrium (Balasubramhanya and Doyle III, 2000).

Reactive distillation is a process that combines both separation and chemical reaction into one unit. It has a lot of advantages for those reactions occurring at temperatures and pressures suitable for the distillation of the resulting components. Its main advantages are derived from the elimination of equipment and the constant removal of products (Sneesby, Tade, Datta, and Smith 1997). It is known that increased overall conversion can be achieved in equilibrium reactions if the products are continuously removed from the reaction zone. Its other advantages include reduced investment and operating costs, environmental impacts (Pérez-Correa, González, and Alvarez 2008), higher conversion, improved selectivity, lower energy consumption, scope for difficult separations and avoidance of azeotropes (Jana and Adari, 2009).

However, due to the occurrence of both reaction and separation in a single unit, reactive distillation exhibits complex behaviors (Khaledi and Young, 2005) such as steady state multiplicity, process gain sign changes (bidirectionality) and strong interactions between process variables (Jana and Adari 2009). These complexities have made the modeling of the process extremely difficult especially when the column type is a packed one and the reaction is solid-catalyzed. Thus, the representation of this process in the form of a model is still a challenge to chemical engineers because the reactive distillation process, especially one involving continuous flows of feeds, is never truly at steady state. Feed and environmental disturbances, reboiler fouling and catalytic degradation continuously upset the conditions of the smooth running of the process. One approach discovered for the representation of the behavior of a process like this (reactive distillation) is the use of Aspen HYSYS process simulator.

Aspen HYSYS is a process simulation environment designed to serve many processing industries. It is an interactive, intuitive, open and extensible program. It also has many add on options to extend its capabilities into specific industries. With this program, rigorous steady state and dynamic models for plant design can be created. Apart from this, monitoring, troubleshooting, operational improvement, business planning and asset management can be performed with the HYSYS simulator. Through the completely interactive HYSYS interface, process variables and unit operation topology can be easily manipulated (Aspen, 2003).

Therefore, this paper is aimed to develop, simulate, optimize and validate, using experimental studies, an esterification process for the production of ethyl acetate using reactive packed distillation column with the aid of Aspen HYSYS.

Procedures

Experimental Procedure

The experimental pilot plant in which the experiments were carried out was a reactive packed distillation column set up as shown in Figures 1a and b. The column had, excluding the condenser and the reboiler, a height of 1.5 m and a diameter of 0.05 m. The column consisted of a cylindrical condenser of diameter and height of 5 and 22.5 cm respectively. The main column section of the plant was divided into three parts of 0.5 m each. The upper, middle and lower sections were the rectifying, the reaction and the stripping sections respectively. The rectifying and the stripping sections were packed with raschig rings while the reaction section was filled with Amberlyst 15 solid catalyst (the catalyst had a surface area of 5300 m^2/kg, a total pore volume of 0.4 cc/g and a density of 610 kg/m^3). The reboiler was spherical in shape and had a volume of 3 Litre. The column was fed with acetic acid at the top (between the rectifying and the reaction sections) while ethanol was fed at the bottom (between the reaction and the stripping sections) with the aid of peristaltic pumps which were operated through a computer via MATLAB/Simulink program. All the signal inputs (reflux ratio (R), feed ratio (F) and reboiler duty (Q)) to the column and the measured outputs (top section temperature (T_{top}), reaction section temperature (T_{rxn}) and bottom section temperature (T_{bot})) from the column were sent and recorded respectively on-line with the aid of MATLAB/Simulink computer program and electronic input-output (I/O) modules that were connected to the equipment and the computer system. At each case of the experimental studies, the operating parameters were fixed based on the conditions being investigated. The reaction taking place in the packed column is given as:

$$CH_3COOH + C_2H_5OH \xleftrightarrow{K_{eq}} CH_3COOC_2H_5 + H_2O \qquad (1)$$

(a) (b)

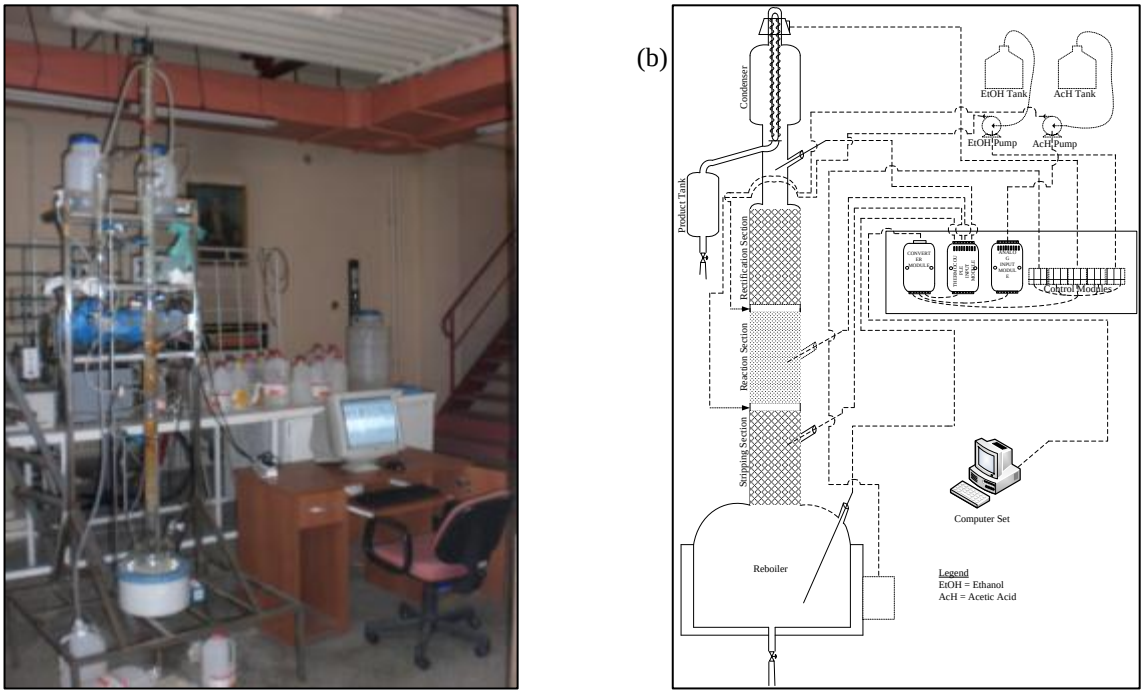

Figure 1 Reactive packed distillation pilot plant: (a) Pictorial view; (b) Sketch view

HYSYS Modeling Procedure

Figure 2 below shows the flowsheet of the reactive packed distillation column built and modeled in HYSYS 3.2 environment. The column consists of a condenser, a rectifying section, an acetic acid feed section, a reaction section, an ethanol feed section, a stripping section and a reboiler. The steady state operating parameters used for the HYSYS model formulation and simulation are as shown in Table 1.

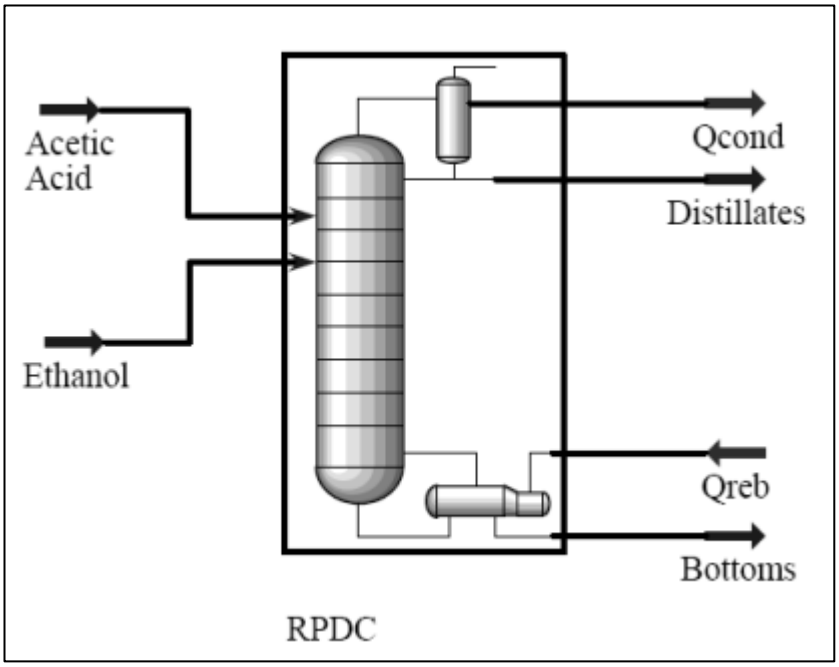

Figure 2 Aspen HYSYS reactive packed distillation steady state simulation flowsheet

Table 1 Steady state operating parameters

Parameter	**Value**
Fluid Package	General NRTL
Reflux ratio (kmol s^{-1} recycle/ kmol s^{-1} distillate)	1
Feed ratio (mL s^{-1} acetic acid/mL s^{-1} ethanol)	1
Reboiler duty (kJ/s)	0.250
Condenser	
Type	Cylindrical
Height (m)	0.225
Diameter (m)	0.05
Rectifying Section	
Packing type	Raschig rings
Height (m)	0.4412
Acetic Acid Feed Section	
Packing type	Raschig rings
Height (m)	0.0882
Reaction Section	
Packing type	Amberlyst 15
Height (m)	0.4412
Ethanol Feed Section	
Packing type	Raschig rings
Height (m)	0.0882
Stripping Section	
Packing type	Raschig rings
Height (m)	0.4412
Reboiler	
Type	Spherical
Volume (L)	3
Level	50%

HYSYS Optimization Procedure

After the steady state simulation, the optimization of the plant was carried using the same HYSYS 3.2 process simulator by incorporating an optimizer into the flowsheet (see Figure 3). Three different algorithms were used for the optimization; they are: Fletcher-Reeves, Quasi-Newton and Successive Quadratic Programming (SQP) algorithms. The objective function of the optimization was taken as maximizing the mole fraction of ethyl acetate in the distillate (top segment) stream. The ranges of the adjusted variables used for the optimizations are as shown in Table 2 below.

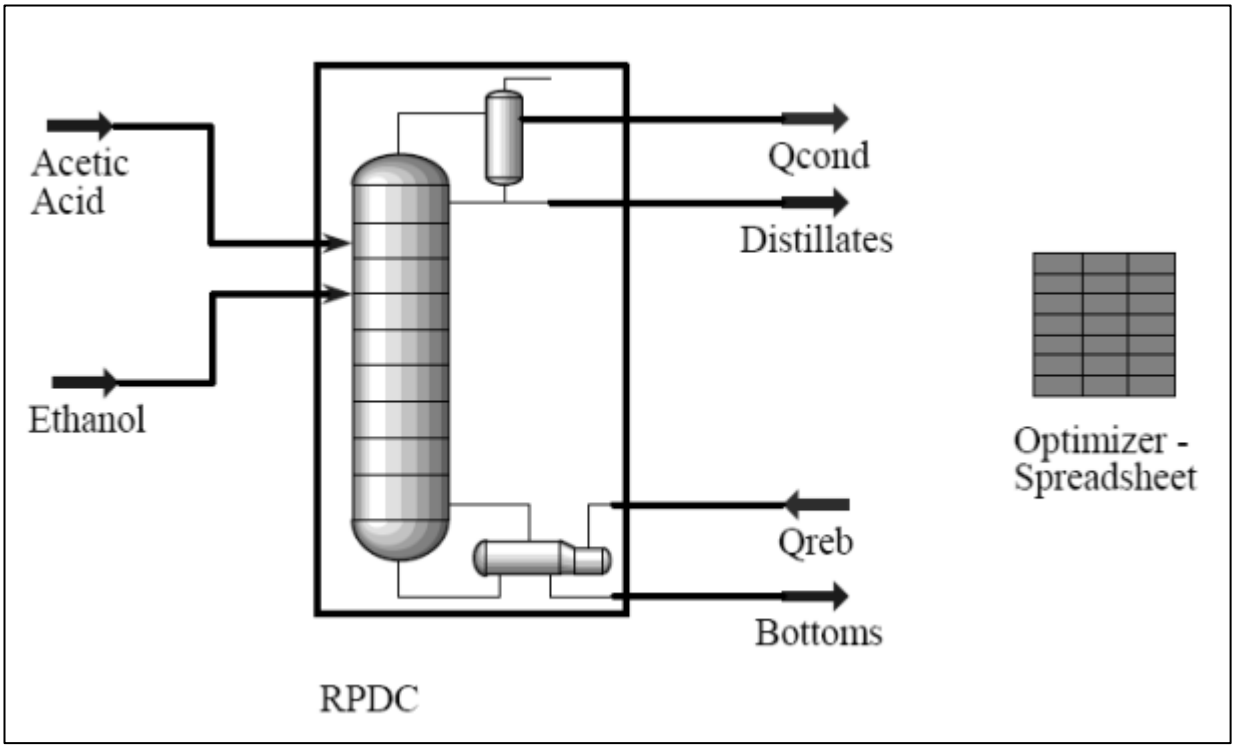

Figure 3 Aspen HYSYS reactive packed distillation optimization flowsheet

Table 2 Parameters used for running the optimizations

Parameter	Low bound	High bound
Reflux ratio (kmol s^{-1} recycled liquid/kmol s^{-1} liquid distillate)	1	9
Feed ratio (mL s^{-1} acetic acid/mL s^{-1} ethanol)	0.5	5
Reboiler duty (kJ/s)	0.050	0.600

After running the HYSYS optimizer, the optimized values of the parameters obtained from one of the algorithms were then used to run the experimental set-up again for validation.

Results and Discussions

To simulate and optimize a reactive packed distillation column for the production of ethyl acetate using Aspen HYSYS 3.2 process simulator in this work, the entıre column was divided into 17 segments excluding the condenser and the reboiler and its steady state study was carried out by simulating the prototype plant built using the simulator under the conditions of reflux ratio of 1, feed ratio of 1 and reboiler duty of 0.250 kJ/s. The other parameters used for the simulation can be found in Table 1. After the simulation, the temperature and composition profiles obtained are as shown in Figures 4 and 5 respectively.

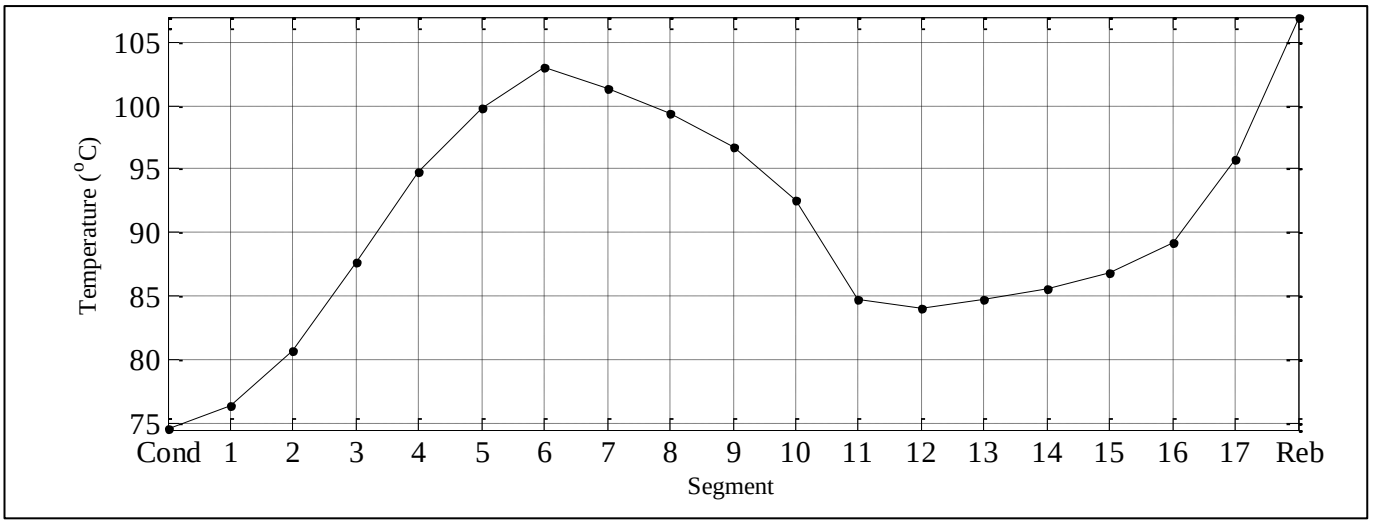

Figure 4 Aspen HYSYS reactive packed distillation column steady state temperature profile

As can be seen from the temperature profile shown in Figure 4, the temperature of the segment near the acetic acid feed section was found to be very high. This was due to the combined effects of the exothermic nature of the reaction occurring in the reaction section of the column, the upward movement of ethanol vapor from the ethanol feed section and the mixed vapor moving upward from the reboiler.

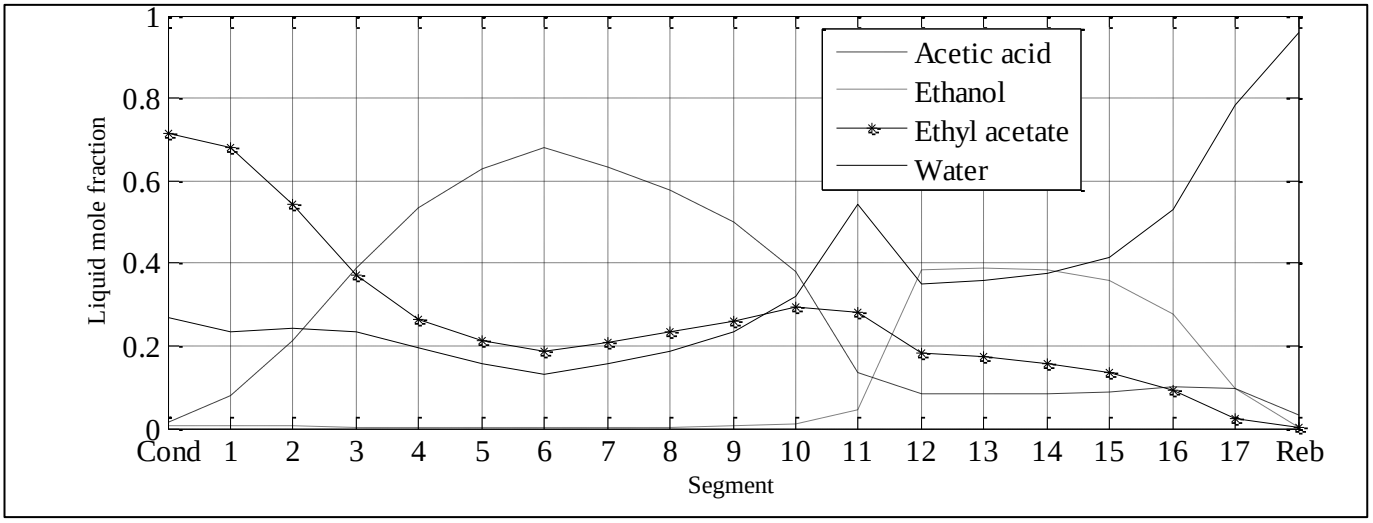

Figure 5 Aspen HYSYS reactive packed distillation column steady state composition profiles

From the composition profile (Figure 5), ethyl acetate (the desired product), as expected, was found to have the highest mole fraction of 0.7132 in the top segment of the column followed by water with a mole fraction value of 0.2665. The mole fractions of the other two components (acetic acid, 0.0141 and ethanol, 0.0062), as expected, were found to be very low in the top segment. This was an indication that effective reaction conversion and separation were achieved in the column.

Considering the mole fractions of the various components in the reboiler, water was found to have the highest mole fraction of 0.9597 followed by acetic acid with mole fraction of 0.0332. The mole fractions of ethanol and ethyl acetate in the reboiler were 0.0043 and 0.0028 respectively. The very small values of the mole fractions of acetic acid and ethanol in the reboiler were due to high reaction conversion occurring in the column.

When the experimental set-up was run using the same parameters as those used for the HYSYS simulation, taking the top temperature as the point of interest to infer composition because it is the one indicating the state and kind of the product obtained from the column, a good relationship was observed from the results of the two (HYSYS simulation and experimental study) cases. For instance, the steady state top segment temperature obtained from HYSYS simulation was 74.5338 ^{o}C while that of the experimental study was measured to be 74.6600 ^{o}C. This is an indication of the fact that the HYSYS model developed for the reactive packed distillation process is a good representation of the real process.

Having carried out the steady state simulation of the Aspen HYSYS RPDC and validated using an experimental study, the process was optimized using three different optimization algorithms. The maximization of the mole fraction of ethyl acetate in the column top segment was set as the objective function of each of the optimizations. The results obtained from the optimizations of the process are as shown in Table 3 below.

Table 3 Optimum parameters

	Value			
Parameter	**Steady-State**	**Fletcher-Reeves**	**Quasi-Newton**	**SQP**
Reflux ratio (kmol s^{-1} recycle/ kmol s^{-1} distillate)	1.0000	2.8995	3.0881	2.6103
Feed ratio (mL s^{-1} acetic acid/mL s^{-1} ethanol)	1.0000	3.3251	3.4565	2.0011
Reboiler duty (kJ/s)	0.2500	0.1076	0.0951	0.1070
Objective function (Top ethyl acetate mole fraction)	0.7132	0.7628	0.7624	0.7608
Top segment temperature (^{o}C)	74.5338	74.3335	74.3356	74.3260

It can be observed from the results shown in Table 3 that the increase (due to the maximization) in the mole fraction of ethyl acetate in the column top segment has resulted in a decrease in the top section temperature. Also, as can be seen from the table, among the three algorithms used for the optimization of the process, Fletcher-Reeves algorithm was found to give the highest mole fraction of ethyl acetate in the top segment of the column by maximizing the objective from the steady state simulation value of 0.7132 to 0.7628. Quasi-Newton algorithm yielded a very very close value (0.7624) of ethyl acetate mole fraction to that of the Fletcher-Reeves algorithm. The optimized ethyl acetate mole fraction (0.7608) gave by SQP algorithm was also found not to be too different beyond acceptation from those of the other two. The differences in the objective functions given by the three algorithms were accounted for by the differences in the optimized operating conditions given by them. For instance, the optimized reflux ratio obtained from the three algorithms can be approximated to one significant figure of 3.

However, the situation is not the same in the case of the feed ratio because the approximations to one significant figure of each of the feed ratios obtained from the Fletcher-Reeves and Quasi-Newton algorithms were both 3 while that of the SQP was 2. Considering the optimum reboiler duty, the values given by Fletcher-Reeves and SQP algorithms were found to be very close to each other than when each of them is compared to the result given by Quasi-Newton.

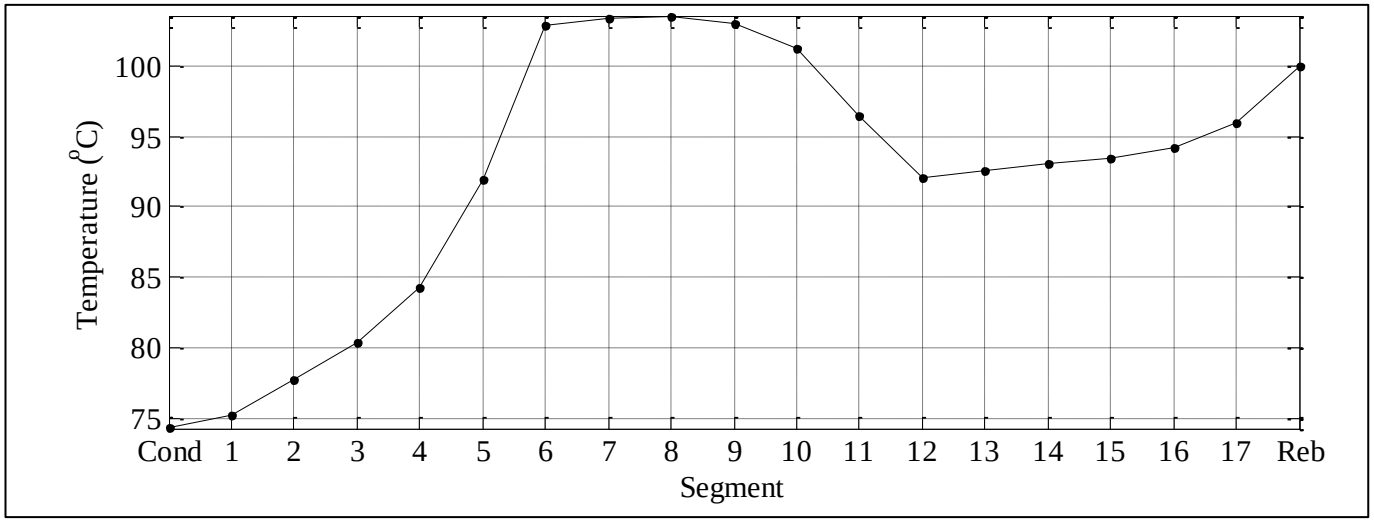

Figure 6 Aspen HYSYS reactive packed distillation column optimization temperature profile

Furthermore, an experiment was carried out for the validation of the optimization using the optimized operating conditions obtained from one of the algorithms. In this case, the optimized operating conditions of SQP was used because the value of its objective function was discovered to be close to those of the other two and it had the lowest feed ratio among the three. Choosing it (SQP) was considered as an effort to reduce cost. After the experiment was carried out using the selected optimum operating conditions, it was discovered from the results that there is a good conformity between the optimized top segment temperature of the process using HYSYS 3.2 and the one measured from the experiment because the HYSYS 3.2 optimized value was 74.3260 ^{o}C while the experimental one was 74.5200 ^{o}C.

From the temperature and composition profiles of the optimized case of the process shown in Figures 6 and 7 respectively, it was noticed that there are changes between the profiles and those of the steady-state simulation shown in Figures 4 and 5. That is to say that, while trying to maximize the composition of the ethyl acetate in the product stream, the changes that occurred in the composition profiles of the components have caused a change in the temperature profile also.

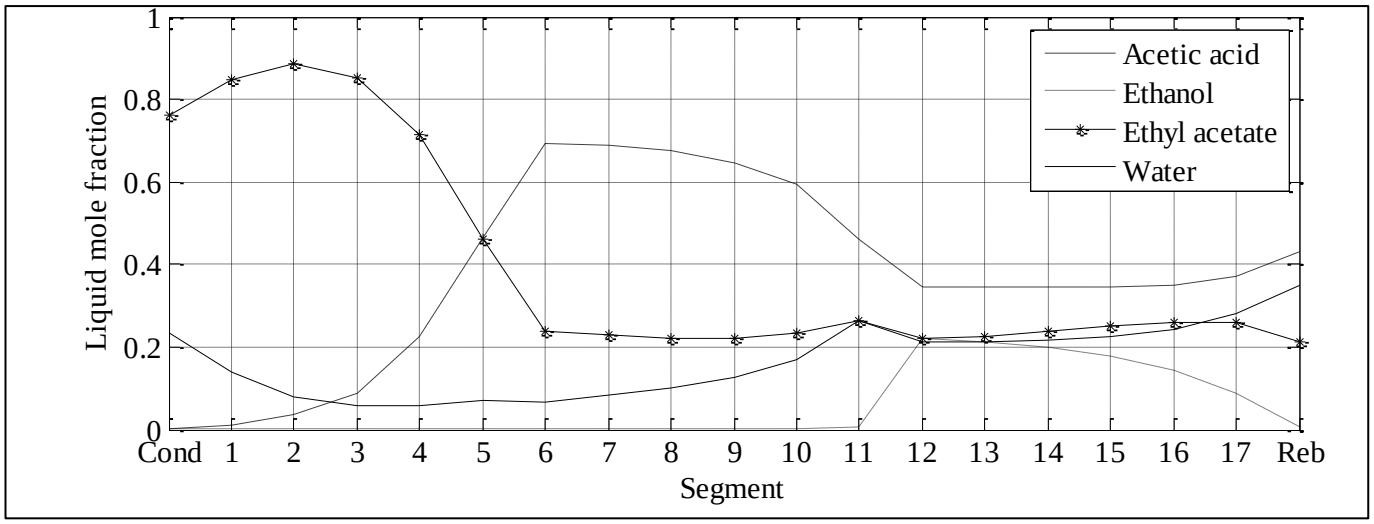

Figure 7 Aspen HYSYS reactive packed distillation column optimization composition profiles

The occurrence of the change in the temperature profile owing to the changes in the composition profiles of the components is an indication of the fact that the compositions of the components and the temperatures of the column segments are dependent on one another. In other words, column segment composition is a function of column segment temperature and vice versa.

Conclusions

The good relationship between the temperature estimated from the simulation using the developed Aspen HYSYS model for the reactive packed distillation process and the experimental ones measured from the pilot plant have revealed that Aspen HYSYS can be used to represent and simulate the process successfully. The three optimization algorithms investigated were found to produce relatively similar maximized mole fractions of ethyl acetate in the top segment of the column. When the optimum parameters of SQP were used to run an experiment for validation, a good agreement was found between the optimum top segment temperature and the experimental one.

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