

# Simulation Analysis of an Industrial Heterogeneous Azeotropic Distillation Column for Control, using operating paths.

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## Abstract

The separation of components constituting an azeotropic mixture cannot be performed with a classical distillation column. One answer to this problem consists in using heterogeneous azeotropic distillation. A separation agent forming an heterogeneous azeotrope with one of the components is added to help to pass through the azeotrope barrier and facilitate the solvent reprocessing, often through the use of a decanter.

A complete study of an industrial heterogeneous azeotropic distillation is presented. This process concerns an organic acid dehydration using an entrainer. Furthermore a fourth component, a by-product is considered.

The study follows a systematic procedure. It includes a validation of the model through a pilot process study, with experimental campaigns, simulation of the pilot and a comparison between experimental and simulation results. The pilot column shows an interesting behavior since it switches from a low temperature profile to a high one for a small variation of boiler heating.

Then a simulation of the industrial process has been made with plate by plate MESH (Material balance, Equilibrium, Summation of fractions and Heat balance) equations. The use of an operating path tool (Alliet-Gaubert et al., 1999) us allows to resolve the convergence difficulties encountered previously. Simulation results are compared with a test run and the matching is good. Furthermore, the operating path shows a process unusual behavior : the temperature of the sensible plate forms a peak according to heat power variation. The sharp variation of the temperature profile of the column and especially the peak of the sensible plate may explicate why it is difficult to control the process on the industrial set point that lie close to the maximum of the peak.

## Scope

This study concerns a running industrial process with a two columns sequence (azeotropic column and solvent recovery column). Four of the main components are considered.

A systematic study procedure which includes a model testing on a pilot column and a case study on the industrial process provides a better understanding of the process operation. Simulation results are very close to the experimental data.

The simulation operating path results show an unusual process behavior and give a valuable explanation of control difficulties that initiated this study.

## Conclusions and significance

This study confirms (as shown by Kowach and Seider, 1987 and Pham et al., 1989) the relationship between impurities in the bottom stream and column temperature profile. It shows as well the connection between a high water fraction and a low temperature in the column.

Results display an unusual peak form response of the sensible plate temperature according to heat power variation. The control difficulties observed on the industrial unit may be caused by the location of the reference temperature close to the maximum of this peak.

This study shows as well the operating path tool interest and the necessity of a systematic study to tackle the operation of real industrial processes.

## ***1. Introduction***

Distillation is a separation technique which uses volatility differences to separate chemical species of a mixture, where "chemical species" are (more or less) pure components or components groups. Homogeneous azeotropic mixtures are non-ideal mixtures characterized by the occurrence of homogeneous azeotropes (same liquid and vapor compositions) and full miscibility in the liquid state. Heterogeneous azeotropic mixtures are non-ideal mixtures characterized by the occurrence of homogeneous or heterogeneous azeotropes (the later being defined by two liquid phases and a single vapor phase which composition equals the overall liquid one) and the presence of an immiscibility region in the liquid state.

The presence of an azeotrope adds some difficulties for separation which use volatility differences. For example, in binary mixtures distillation, the azeotrope is obtained either at the top or at the bottom of the column and sets an impassable barrier keeping from obtaining both pure components with a single column at given operating conditions.

In order to separate azeotropic mixtures several technologies may be used:

- 1- Pressure swing distillation consists in two columns which pressure difference gives the ability to overcome the azeotropic barrier.
- 2- In homogeneous azeotropic distillation, a third component is added so as to modify the components relative volatility.
- 3- Heterogeneous azeotropic distillation is based on the same principle as homogeneous azeotropic distillation, but the added third component is partially miscible with one of the components, Solvent reprocessing is easy by means of a liquid-liquid separation system.
- 4- Reactive distillation is based on the transformation of one of the components into a component which does not form an azeotrope with the other components.
- 5- Salted distillation consists in adding a ionic salt that dissociates in the liquid mixture and changes the azeotrope composition.

Heterogeneous azeotropic distillation (HAD) is often preferred in industry because it offers a good economic compromise due to easy separation of the two liquid phases in a decanter. However liquid - liquid demixions may arise onto the plates and make both behavior understanding and process control difficult.

In this article, a complete study of an industrial process is presented. Its aim was to explain industrial process operation and control difficulties leading to purity requirements unmet. First, difficulties commonly encountered in heterogeneous azeotropic distillation are reminded. Second, the studied process is described and results of former studies are presented. A third part insists on modeling aspects used in this study. The resulting model is validated, in the fourth part by the simulation of the pilot unit. Then, the industrial process operation is simulated and explanation to the control difficulties encountered in industrial practice are discussed.

## ***2. Difficulties of heterogeneous azeotropic distillation***

Widagdo and Seider (1996) have reviewed azeotropic distillation and in particular the key features of heterogeneous azeotropic distillation. We extracted the following difficulties relative to heterogeneous azeotropic distillation:

- The design offers a lot of possibilities in the choice of a separation agent. They extend the

conceivable combinations and make the design more difficult to be performed efficiently.

- The column behavior is not well known. An explanation may be the occurrence of steady state multiplicity which can be classified into three classes, (Esbjerg et al, 1998) namely multiple input, multiple steady-state or multiple output.
- Since the behavior is not well understood, the control of such columns is very difficult and is based on practitioners know-how.

On the industrial unit, practice has shown that some high fluctuations in the temperature profile may happen and are related to variations of impurities concentrations in the bottom stream. That is why the control of this process is both very subtle and very important. Several steady-state simulations of the process were made and up to now they were not able to represent the process behavior clearly. Three explanations seemed interesting to explore :

- The process behavior oscillates between multiple outputs.
- The process operation presented as a steady-state is not, in fact, a true stationary operation and is maintained by a control loop.
- The causes of this behavior are hydrodynamic with phenomena of raw hit type. This behavior can be represented only by a hydrodynamic modeling.

A systematic method consists in exploring these assumptions in the order of work easiness which is the order of appearance. Multiple output may be found with steady-state simulations with an operating path tool (Alliet-Gaubert et al., 1999). Control maintenance state needs a dynamic simulator. The third assumption study requires a Computer Fluid Dynamic simulator. This study addresses the first explanation.

### ***3. Description of the industrial process unit and of the pilot unit***

#### *3.1. Industrial unit*

Very strict confidential agreement prevents us to describe in extenso the process unit, in particular the name of the product, of the main impurity and of the entrainer solvent, hereafter called PRODUCT, BY-PRODUCT and SOLVENT. As a matter of fact, we are dealing in this study with a quaternary heteroazeotropic system.

The purpose of the studied process is the dehydration of an organic acid. Since boiling and condensing temperatures of these two chemical species have the particularity of forming a pinch close to the pure Water locus (Figure 1), usual distillation is not convenient to recover all the PRODUCT. The industrial process consists in adding an organic SOLVENT that makes a heterogeneous azeotrope with WATER.

The industrial process studied consists in a 52 plates (including boiler and decanter) azeotropic distillation column C1 and a 29 plates (including boiler and decanter) solvent regenerating column C2 (see Figure 2). The pure PRODUCT is collected in the bottom stream of the C1 column and the low boiling heterogeneous azeotropic mixture of WATER and SOLVENT exits at the top of C1. This later stream is fed into a decanter. The aqueous phase is driven to the solvent regenerating column C2 where pure WATER is collected at the bottom and SOLVENT is recycled to the decanter. The organic phase is refluxed to the azeotropic tower. A small auxiliary stream of SOLVENT is necessary to replace small losses in the C1 bottom PRODUCT and the C2 bottom WATER streams.

A previous study (Letourneau 1994) has shown the relation between slight variations of WATER and SOLVENT concentrations in the C1 bottom output and big operating differences substantiated through large plate temperature changes. The same kind of problem have been found for different

components by Kowach and Seider (1987) and Pham et al. (1989).

### **Figure 1: Liquid-Vapor equilibrium curves for WATER-PRODUCT**

#### *3.2. Pilot unit*

Few measures are readily obtained on the industrial process, making the assessment of the validity of the simulation model difficult. Therefore, a pilot unit has been built to acquire the necessary information needed to test the simulation model. The pilot unit is composed of the azeotropic tower alone with 18 plates, a boiler and a decanter (Figure 3). Input and output streams are connected to stock tanks. For technical reasons, the organic reflux is not directly refluxed in the column but is conveyed through a stock tank.

### **Figure 2: Industrial unit**

### **Figure 3: Pilot unit structure and experimental data Measurements and control system description.**

#### *3.3. Industrial unit*

A test run of the process provides:

- stream flowrates and compositions through the whole process,
- temperatures of plates number 1, (decanter) 2, 9, 18, 24, 30, 38, 42, 51, 52 (boiler) of the C1 column and temperature of plates 1, 11, 21, 29 (boiler) of the C2 column,
- top and bottom pressure of columns C1 and C2, pressure of the decanter,

The accuracy of those test-run experimental values is not assessed and a hand-made data reconciliation was made so as to satisfy material balances.

Control of the process aims at keeping SOLVENT or WATER content in the PRODUCT stream under a few hundreds ppm to meet the PRODUCT purity requirements.

For each column, the control loop consists in a measurement of the temperature of a sensitive plate and an action accordingly on the boiler heating. Sensitive plate numbers are 42 for C1 and 21 for C2.

#### *3.4. Pilot measurements and control system description*

For the pilot unit, compositions and flow rates of the three input streams and of the three output streams are known. Temperatures of plates number 1 (decanter), 2, 6, 10, 14, 18 and 20 (boiler) are measured. Experimental input and output streams compositions and flow rates are reconciled to satisfy partial and total material balances. An example of typical reconciled input and output flowrates and compositions is shown on Figure 3.

### **4. Process modeling and simulation**

The simulation study is done with a case study tool called ProCase (Alliet-Gaubert et al., 1999). This tool is able to follow an operating path and to restart from an operating path direction. An operating path is a succession of steady states, that is characterized by continuously derivable output results; it gives a good idea of how the process behaves during a slow variation of an operating parameter. It can also detect multiple solutions that belong to different operating paths.

Convergence and operating paths are all the more reached since each steady state solution is obtained using for the initializing conditions the previously converged steady state results.

The column model (ProSim 1993) consists of usual plate by plate MESH (Material balance, Equilibrium, Summation of fractions and Heat balance) equations which are solved for the whole column, decanter included. A first calculation is done with liquid-vapor equilibrium (LVE) equation for each plate. Then, if demixion is detected for first calculated compositions and temperature, a new calculation is made with liquid-liquid-vapor equilibrium (LLVE) equations for the triphasic plates and LVE equations for others plates. Since spurious multiple steady states can appear because of too loose convergence criteria (Rovaglio et al, 1993), tight convergence criteria values of  $10^{-9}$  are set.

LVE and LLVE are described by the NRTL thermodynamic model with six binary coefficients (Renon 1971). Possible dimerisation of the PRODUCT and the BY-PRODUCT in the vapor phase is taken into account by the model of "Vapor Phase Association" (ProSim 1993). Binary coefficients have been identified on experimental data at the appropriate pressures. These coefficients have been checked to give good representation of binary mixtures (for example Figure 1) and we have checked graphically that no false ternary azeotrope was created.

## 5. Pilot unit simulation

### 5.1. Description of the simulation

The column functioning is assessed using a steady-state simulator in combination with a case study tool. Fluctuations of the organic phase quantity in the stock tank can not be represented in the steady-state way. Consequently, it is represented by a stream which direction (input or output) depends on the variation direction. For physical and convergence reasons, input parameters are the reflux molar flowrate and the boiler heating. We do not know the boiler heating due to heat losses during the transportation. This enforced the interest to use this variable as a command parameter for the sensitivity analysis performed with the case study tool. Plate efficiencies are 0.5 for the top of the column and 0.7 for the bottom, in accordance with literature value of the plate type used. A typical simulation is presented in the Figure 4.

### 5.2. Comparison of experimental results with simulation ones

A range of boiler heating value from 650 to 700 kcal/hr is covered at constant molar reflux flowrate and resultant steady state solutions are analyzed.

Temperature profiles in the column for various boiler heating are shown on Figure 5. The agreement between the experimental and simulated temperature is good on the feed plate and on the boiler (The agreement is good on the condenser because the temperature is fixed). With boiler heating variation, the temperature is varying on the lower part of the column and is passing through the experimental value whereas the temperature is not changing on the upper part of the column.

For a boiler heating of 658.5 kcal/hr which corresponds to the best temperature profile accordance (see Figure 5), experimental and simulated concentrations and flowrates of output streams are presented in Table 1. As the low error values show, experimental and simulated concentrations and flowrates are in good agreement, especially if we consider the modeling approximation which converts an accumulation to an output flowrate (organic distillate).

### Figure 4: Pilot unit simulation

Figure 5 and Figure 6 display respectively the temperature profile for three boiler heating values and the variation of the sensitive plate temperature and of the SOLVENT and WATER content in the

PRODUCT-rich column bottom stream with boiler heating value. We notice on Figure 5 that:

- 1- two types of temperature profiles in the column are seen, a low one for boiler heating lower than 657 kcal/hr and a high one for boiler heating higher than 660 kcal/hr,
- 2- the shift between these two profiles is sharp as the temperature of the sensitive plate may vary of 25 °C for a 4 kcal/hr (0.6%) variation of the boiler heating.

		Expement.	Simulated	Error
QB			658,5	
Org. dist.	Water	0,033	0,044	0,011
	PRODUCT	0,035	0,037	0,002
	BY-PRODUCT	0,028	0,018	-0,010
	SOLVENT	0,904	0,901	-0,003
	florate (g/h)	170,9	170,5	-0,2%
Aqu. dist.	Water	0,938	0,942	0,004
	PRODUCT	0,012	0,010	-0,002
	BY-PRODUCT	0,030	0,027	-0,003
	SOLVENT	0,020	0,021	0,001
	florate (g/h)	660,0	656,1	-0,6%
Bottom	Water	7,00E-04	3,35E-06	-6,97E-04
	PRODUCT	0,989	0,987	-0,002
	BY-PRODUCT	0,010	0,013	0,003
	SOLVENT	2,00E-04	4,07E-05	-1,60E-04
	florate (g/h)	1125,4	1129,7	0,4%

**Table 1 : Comparison of experimental and simulated output for  $Q_b = 658.5$  kcal/hr**

The discrepancy of the experimental and simulated temperature profile on the upper part of the column is not a very important concern as the pilot simulation aims at validating both the plate by plate MESH steady-state simulation model and liquid-liquid-vapor equilibrium (thermodynamic) model in terms of physically coherent results. What is more interesting is the behavior shown on Figure 6.

**Figure 5: Pilot temperature profile for various boiler heating**

**Figure 6: Plate 14 temperature and bottom impurities compositions variations with boiler heating.**

**Figure 7: Variations of the recovery rates in the bottom product with boiler heating.**

**Figure 8: Variation of molar concentrations in the column for  $Q_b=657$  kcal/hr.**

**Figure 9: Variation of molar concentrations in the column for  $Q_b=658.5$  kcal/hr.**

**Figure 10: Variation of molar concentrations in the column for  $Q_b=660$  kcal/hr.**

### 5.3. Pilot column behavior

Around the reference temperature, a tiny variation of the boiler heating command parameter makes an important variation of the temperature of the sensitive plate n°14. This readily explains its choice as a control measure.

The sharp temperature increase is accompanied by a steep decrease of the WATER content in the bottom stream (Figure 6), whereas SOLVENT content first increases up to a concentration maximum and then diminishes.

The sensitive plate temperature value chosen corresponds to an impurity concentration minimum in the PRODUCT-rich bottom stream. Interestingly, the process engineers operated the pilot unit so as to minimize the impurities in the PRODUCT-rich stream.

Operating at high  $Q_b$  value with a high temperature profile seems acceptable on the basis of impurity content in the PRODUCT-rich bottom stream. Unfortunately it greatly impairs the PRODUCT and BY-PRODUCT mass yield as shown on Figure 7. Indeed, the PRODUCT recovery value decreases and falls under the 95% weight specification value when  $Q_b > 665$  kcal/hr.

The almost perfect symmetrical variations of the BY-PRODUCT recovery value and temperature of the plate 14 temperature is a coincidence (see Figure 7), since the same simulation procedure made without BY-PRODUCT gives a similar abrupt temperature profile (see Figure 11).

This phenomena gives us some insight on the process. When looking at the mass liquid fraction profiles, (see Figure 8 to Figure 10), we noticed that as  $Q_b$  goes up, WATER is removed from the bottom stream. Once the bottom stream is almost depleted from WATER, the BY-PRODUCT starts moving up the column, followed by the PRODUCT.

The temperature front on plate 14 is associated with concentration fronts moving along the column as seen on Figure 9. As  $Q_b$  increases, a BY-PRODUCT concentration maximum moves up, pushing a WATER concentration front up the column. Bottom stream is richer in PRODUCT but its flowrate decreases, inducing a yield loss.

We notice also on Figure 6 that the number of heterogeneous plates varies from 4 when the temperature profile is low to 1 (decanter) for the high temperature profile. The increasing presence of BY-PRODUCT and PRODUCT on the upper plates set the liquid mixture away from the demixion zone.

As a conclusion for the pilot unit simulation, both simulation and thermodynamic models enables us to find a temperature on a sensitive plate that corresponds to an impurity concentration minimum in the PRODUCT-rich bottom stream. This value is identical to the one set during the pilot unit operation. We observe a high and a low temperature profile with impurities concentration inversion. Impurity content in the bottom stream affects strongly the temperatures in the lower part of the column.

Those results not only fully validate the simulation and thermodynamic model that are used for the simulation of the industrial unit, but also give information of what may be expected for the simulation of the industrial installation.

**Figure 11: Plate 14 temperature and bottom impurities compositions variations with boiler heating, simulation without BY-PRODUCT.**

## 6. Process simulation

### 6.1. Description of the simulation study

The simulation of columns C1 and C2 together is similar to the industrial structure (see Figure 2). The efficiency is taken to 0.3. Since the liquid-liquid separation determines automatically the reflux flowrates, only two input parameters are necessary. In the industrial process, boilers heating are used to control the temperature of the reference plate for each column. That is why the two boilers heating are the simulation input parameters of which variations influence is studied.

	Feed	Stab1	Stab2	Solvent
Water	0.315	0.447	0.666	0.0
PRODUCT	0.662	0.553	0.334	0.0
BY-PRODUCT	0.023	0.0	0.0	0.0
SOLVENT	0.0	0.0	0.0	1.0
Flowrate (kg/hr)	45135.	381.	717.	To balance losses

**Table 2: Feeds compositions and flowrates**

We followed an operating path procedure with C1 boiler heating ( $Q_{b1}$ ) varying from 12750000 kcal/hr to 17354447 kcal/hr. For higher values we could not manage to get simulation convergence on the same operating path. Already from  $Q_{b1} = 17250000$  kcal/hr to  $Q_{b1} = 17354447$  kcal/hr, the convergence is difficult.

We tried to follow an operating path with  $Q_{b2}$  as an operating parameter, but it proved to be a bad choice because of the behavior type as described in the next paragraph. So we followed operating path with  $Q_{b1}$  as a parameter for various  $Q_{b2}$ .

### 6.2. Column behavior

We notice the same type of behavior that the pilot column one: when the sensible plate temperature is equal to the reference value, the total amount of impurities (Water and SOLVENT) in the Bottom1 stream is minimum (see Figure 12).

The water contents has the same influence on the temperature profile: a big amount of water corresponds to a low temperature profile.

However, three observations are different from the pilot study : (i) the variation of the column C1 sensible plate temperature with  $Q_{b1}$  does not form a plate but a peak ; (ii) there seems to be some convergence difficulties when  $Q_{b1}$  is superior to the  $Q_{b1}$  corresponding to the peak, (iii) the number of triphasic plates grows with  $Q_{b1}$  instead of diminishing.

Moreover, for different  $Q_{b2}$ , the variation of the sensible plate temperature with  $Q_{b1}$  has the same shape. However, the  $Q_{b1}$  value corresponding to the peak and the temperature maximum of the peak are varying with  $Q_{b2}$ .

### 6.3. Comparison of experimental and simulated results

We manage to get a couple ( $Q_{b1}$ ,  $Q_{b2}$ ) which gives good agreement not only for the temperature profile as shown on Figure 13, but also for output streams compositions and flowrates as shown on Table 3.

<i>Mass fractions in RESIDU1</i>			<i>Mass fractions in RESIDU2</i>		
	<i>TEST RUN</i>	<i>simulated</i>		<i>TEST RUN</i>	<i>simulated</i>
<i>PRODUCT</i>	96,8%	96,7%	<i>PRODUCT</i>	2000 ppm	1518 ppm
<i>Water</i>	2586 ppm	2,7 ppm	<i>Water</i>	98,6%	99,45%
<i>BY-PRODUCT</i>	2,88%	3,2%	<i>BY-PRODUCT</i>	8000 ppm	2998 ppm
<i>SOLVENT</i>	52 ppm	4,6 ppm	<i>SOLVENT</i>	91 ppm	-
<i>Flowrates C2203(kg/hr)</i>			<i>Flowrates C2202 (kg/hr)</i>		
	<i>TEST RUN</i>	<i>simulated</i>		<i>TEST RUN</i>	<i>simulated</i>
<i>Distillate1</i>	72803	81523	<i>Distillate2</i>	708	509
<i>Reflux1</i>	57999	66664	<i>Reflux2</i>	25036	25538
<i>RESIDU1</i>	31312	31373	<i>RESIDU2</i>	24428	24343

**Table 3: Comparison of compositions and flowrates**

#### 6.4. Control difficulties explanation

Since functioning Qb1 is close to the peak maximum, these results leads to the comment that it may be difficult for the controller to hold on the set point: if Qb1 decreases, the temperature of sensible plate decreases and if Qb1 increases, the temperature decreases as well. Further more, Qb2 variations change the peak Qb1 value which enforce the control difficulty.

**Figure 12: Sensible plate temperature and bottom impurities compositions variations with column C1 boiler heating Qb1.**

**Figure 13: Experimental and simulated temperature profiles**

#### Conclusion

This study leads to:

- The validation of the simulation models.
- Process unstability explanations, that the industrial partner was not able to untangle for several years.

Since we were able to find the simulation point we were looking for and which reveal to be very difficult to get, this study shows the interest of the tool ProCase.

We completed this study because of:

- 1- a systematic verification of the modelisation
- 2- a good modeling and calculations
- 3- an adapted tool for convergence and understanding an installation behavior.

The next steps are sensibility analysis and dynamic simulation in order to get a control policy development.

#### Notations

- $D_{mas}, D_{mol}$  mass, molar flowrate  
 $x_i$  molar fraction of component i  
 Qb Heat power

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