Batch distillation of binary mixtures: preliminary analysis of optimal control
Marija Stojkovic, Vincent Gerbaud, Nataliya Shcherbakova

To cite this version:
Marija Stojkovic, Vincent Gerbaud, Nataliya Shcherbakova. Batch distillation of binary mixtures: preliminary analysis of optimal control. IFAC-PapersOnLine, Elsevier, 2017, 20th IFAC World Congress, 50 (1), pp.4899-4904. <10.1016/j.ifacol.2017.08.743>. <hal-01714049>

HAL Id: hal-01714049
https://hal.archives-ouvertes.fr/hal-01714049
Submitted on 21 Feb 2018

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L’archive ouverte pluridisciplinaire HAL, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d’enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.
Open Archive TOULOUSE Archive Ouverte (OATAO)

OATAO is an open access repository that collects the work of Toulouse researchers and makes it freely available over the web where possible.

This is an author-deposited version published in: http://oatao.univ-toulouse.fr/
Eprints ID: 19563

To link to this article: DOI: 10.1016/j.ifacol.2017.08.743
URL: https://doi.org/10.1016/j.ifacol.2017.08.743

To cite this version:
Stojkovic, Marija and Gerbaud, Vincent and Shcherbakova, Nataliya. Batch distillation of binary mixtures: preliminary analysis of optimal control. (2017) IFAC-PapersOnLine, vol. 50 (n° 1). pp. 4899-4904. ISSN 2405-8963

Any correspondence concerning this service should be sent to the repository administrator: staff-oatao@listes-diff.inp-toulouse.fr
Batch distillation of binary mixtures: preliminary analysis of optimal control

Marija Stojkovic* Vincent Gerbaud** Nataliya Shcherbakova***

* Laboratoire de Génie Chimique, Université de Toulouse, CNRS, INPT, UPS, Toulouse, France (e-mail: marija.stojkovic@ensiacet.fr)
** Laboratoire de Génie Chimique, Université de Toulouse, CNRS, INPT, UPS, Toulouse, France (e-mail: vincent.gerbaud@ensiacet.fr)
*** Laboratoire de Génie Chimique, Université de Toulouse, CNRS, INPT, UPS, Toulouse, France (e-mail: nshcherbc@ensiacet.fr)

Abstract: The present work concerns the distillation of a binary non-ideal zeotropic mixture in an N-stage batch column under total condensation and constant vapour flow rate assumptions. The aim is to maximize the final product of desired purity using the reflux as a control parameter. We discuss certain methodological aspects of the numerical resolution, and present the first results obtained by the application of the direct method based on a full discretization of the optimal control problem.

Keywords: Batch distillation, optimal control, maximum distillate problem, direct method

1. INTRODUCTION

Distillation is the technique of separation of the components of a liquid mixture using the principle of relative volatility and different boiling temperatures of the components. It is the most widely used separation technique in chemical industry. In simple distillation, the liquid mixture is kept boiling in a still under thermodynamic equilibrium conditions. The vapour produced along the process is richer in light components than the liquid, it is evacuated from the still by condensation allowing to obtain the distillate rich in desired light component. The process can be intensified by reversing the part of the distillate back to the still, i.e., by using the reflux, which makes the transfer of lighter components to the vapour phase more efficient.

In the industrial context the process is carried out in continuous, batch, or semi-batch distillation mode in distillation columns. The batch distillation is used when a small amount of product of high quality is required, or the flexibility of the production is needed. The most commonly used control parameters are the reflux policy and the reboiler heat duty.

The present paper focuses on the distillation of homogeneous non-ideal binary mixtures. Different strategies were proposed to optimize this process by choosing the best step-wise constant reflux ratio (Farhat et al., 1991), by selecting an optimized repeating sequence of zero and total reflux periods (cyclic operation, Sorensen (1999)), etc. Optimal control of batch distillation of binary mixtures was analysed in minimum time, maximum distillate or maximum economical profit formulations (Mujtaba, 2004). In particular in Converse, Gross (1963) and then in Diwekar (1992), Kim et al. (2001) Potryagin’s maximum principle (PMP) was applied to solve the maximum distillate problem. The authors considered the dynamics on the internal part of the column in almost steady-state approximation. The singular type control policy found by the authors is now considered as the reference optimal solution of the problem. In our opinion certain optimal solutions were neglected in this studies.

We consider in this paper the problem of maximisation of production of the distillate of prescribed purity over a fixed interval of time by controlling the distillate rate. In Section 2 we analyse the mathematical model of the problem. We show that the purity constraint can be transformed into an additional state of the system. Then the maximum distillate problem can be formulated as a standard affine control problem with bounded control and simple state constraints. According to PMP, optimal controls can be both of bang and singular types. In Section 3 we present the results of the numerical resolution for three binary mixtures in batch columns with different number of plates. The presented solutions were obtained by the direct method implemented in the BOCOP solver (Bonans et al. (2014)). We show that the optimal control structure depends on the difficulty of the separation task and on the column configuration. In certain cases the optimal control is almost of bang-bang type, which is very close to the cyclic operation mode reported by other authors. We test one of these optimal scenarios in dynamical simulation using ProSim software (ProSim, 2000), and found a significant amelioration with respect to the conventional optimal batch operation.
2. OPTIMAL CONTROL OF BATCH DISTILLATION

2.1 Dynamical model of batch distillation

The simple batch mode configuration considered in this paper is shown in Fig. 1. It consists of a reboiler, a number of intermediate plates (or trays), used to bring the vapor and liquid phases into contact to enhance the mass transfer, a condenser, and an accumulator tank containing the distillate. For a given mixture, in order to assure the feasibility of the production of the distillate of a specified quality, the number of plates (including the reboiler and the condenser) must be greater than $N_{min}$. This number can be computed in relation to the initial composition and the desired purity of final product via the standard McCabe – Thiele method (Doherty, Malone (2001)). The greater the difference in relative volatility of the components of the mixture, the easier is the separation task.

To describe the thermodynamic equilibrium between the vapor and the liquid phases we use the generalized constant relative volatility model due to Gmehling (Doherty, Malone (2001)):

$$y(x) = \frac{ax}{1 + (a - 1)x} + bx(1 - b).$$

(1)

We consider a batch distillation column under assumption of equinomar overflow and total condensation. The vapor hold-up as well as the pressure drop along the column are neglected, the liquid and the vapor phases are supposed to be perfectly mixed on the plate. These assumptions imply that the values $U_i^*$ and $U_i^0$, $i = 2, \ldots, N - 1$, representing the molar hold-ups for condenser and internal plates are constant, so the complete state space of the N-plates column can be defined as

$$X_0 = \{ q = (x_0, x_1, \ldots, x_N, U_0, U_N) : x_i \in [0, 1] \forall i \}.$$

Here $x_0, x_1, \ldots, x_N$ are molar concentrations of the light component in the liquid phase in accumulator, condenser, intermediate plates and the reboiler correspondingly, while $U_0$ and $U_N$ are the liquid hold-up in the accumulator tank and in the reboiler. Below we mark with the superscript "on" the initial values of these variables. The mass balances over the column yield the system of differential equations (Mujtaba (2004)):

$$\begin{align*}
\frac{dx_0}{dt} &= \frac{u}{U_0}(x_1 - x_0), \\
\frac{dx_i}{dt} &= \frac{1}{U_i}(L_i(x_{i-1} - x_i) + V_i(y_{i+1} - y_i)), i = 2, \ldots, N - 1, \\
\frac{dx_N}{dt} &= \frac{1}{U_N}(L(x_{N-1} - x_N) - V(y_N - x_N)),
\end{align*}$$

$$\frac{dU_0}{dt} = u, \quad \frac{dU_N}{dt} = -u.$$  

(2)

The value $V = const$ is the constant vapor rate between the plates of the column, $L$ and $u$ are variable liquid and distillate rates, and $u(t) + L(t) = V$ for all $t$. These values define the reflux ratio $R = L/u$. The total mass and the mass of the light component are preserved by (2). Note that these equations blow up if either the reboiler or the accumulator tank is empty. To avoid this difficulty we assume that at the beginning of the process the accumulator contains a small amount of distillate of the same concentrations as in the condenser (the first drop assumption).

2.2 Maximum distillate problem

Equations (2) define a control system where $u \in [0, V]$ plays the role of the control parameter. In this paper we consider the problem of maximization of the distillate production of desired purity $y_*$ over in fixed interval of time $t_f$. The standard form of the purity constraint is the following (Converse, Gross (1963), Kim et al. (2001), Uperti (2011)):

$$y_* \int_{0}^{t_f} u(t) dt = \int_{0}^{t_f} u(t)x_1(t)dt.$$  

(3)

Note that $y_*$ describe the purity of the liquid accumulated during the distillation process. The true concentration of the distillate depends on the concentration $x^0_0$ and the amount $U^0_0$ of the first drop of the distillate:

$$x_D = x^0_0 = y_* + \frac{(x^0_0 - y_*)U^0_0}{U_0(t_f)}.$$

In general, $x_D > y_*$, provided $y_*$ is smaller than $x^0_0$, which is always the case for the infinite reflux steady-state initial conditions considered below.

By introducing a new auxiliary state variable $z$ (the purity deviation) verifying

$$\frac{dz}{dt} = u(t)(x_1(t) - y_*), \quad z(0) = 0$$  

(4)

the purity constraint (3) reduces to a terminal time condition $z(t_f) = 0$.

Assuming that at $t = 0$ the column was at steady state under infinite reflux ($u = 0$ for $t < 0$), the maximum distillate problem formulates as...
where the $N + 4$ state variables $U_0, U_N, x_0, \ldots, x_N$ and $z$ verify differential equations (2), (4) and the $(N + 4)$ steady state initial conditions at $t = 0$, as well as the final condition $z(t_f) = 0$. The infinite reflux steady state conditions can be easily computed via recurrence formulae: 
\[ x_i^0 = y_{i+1}(x_{i+1}^0), \quad i = N - 1, \ldots, 1, \]
assuming that the initial concentration in reboiler $x_N^0$ is known.

Due to the mass conservation property of equations (2), the dimension of the state space can be reduced by two constants $m^o = U_N^0 + U_0^0$, $m_x = \sum_{i=0}^{N} x_i^0 U_i^0$, we restrict the original problem (5) to the level sets $U_N(t) + U_0(t) = m^o$, $m_x(t) = m_x^o$, obtaining the reduced state space
\[ X_1 = \{ q = (x_1, \ldots, x_N, U_N, z) : x_i \in [0, 1] \forall i \}. \]

After reduction, the problem can be formulated as the following optimal control problem:
\[
\max_{u(\cdot) \in [0, V]} U_0(t_f), \tag{5}
\]

where $U_0(t_f)$ is the objective function of the problem. If $z(0) = 0$, then $U_0(t_f)$ is a monotone increasing (resp. decreasing) function if $p_{x_0}(0) > 0$ (resp. $p_{x_0}(0) < 0$). On the other hand, the final values $x_0(t_f)$ and $U_0(t_f)$ are non prescribed, the transversality condition of PMP implies $p_{x_0}(t_f) = 0$, $p_{U_0}(t_f) = 0$. Therefore necessarily $p_{x_0}(t_f) = 0$ and hence $p_{U_0} \equiv 0$. So, for the switching function we have $H^{full}_f = H_1$. Moreover, it is easy to verify that \{(0), H^{full}_1\} = \{(0), H_1\}$, so the singular control is of the same form as in the reduced case.

It is worth to remark that several attempts were made in the past (Converse, Gross, 1963), (Dieweke, 1992) to apply the PMP to solve the maximum distillate problem for binary mixtures taking the reflux ratio $R = \frac{V来说}{V + 1}$, $R \in [0, +\infty)$, as the control parameter. The quasi steady-state approximation was used to describe the dynamics in the inner plates. Since $u = V来说 + 1$, the case $R = 0$ corresponds to $u = V$ and $R = +\infty$ (infinite reflux) to $u = 0$. In contrast with the distillate rate $u$, which can be easily implemented in practice, and which leads to a standard optimal control formulation, the use of $R$ has serious disadvantages: the control system (2) is non-linear with respect to $R$, and $R$ is unbounded from above though in reality the value $u = 0$ (infinite reflux) is attained.

Indeed, the cited above authors found only the optimal control of singular type.

3. NUMERICAL RESULTS

Due to the dimension and high non-linearity of the problem, we first solve it numerically. To this end, we consider three homogeneous non-azeotropic binary mixtures, whose thermodynamic properties are well in accordance with the thermodynamic model (1).

3.1 The cases studied

| Light component | Heavy component | a   | b   | $N_{\text{min}}$ |
|-----------------|-----------------|-----|-----|-----------------|
| hexane          | p-xylene        | 7   | 0   | 3               |
| methanol        | water           | 7.15| -0.33| 4               |
| benzene         | ethylenediamine | 9   | -0.6| 6               |
Fig. 2. Liquid-Vapour equilibrium curves for hexane - p-xylene (dashed), methanol - water (dotted), benzene - ethylendiamine (full)

Initially the reboiler is charged with $U_N = 10(mol)$ of liquid, whether each plate (including the condenser and the accumulator tank) contains $U_i = 0.1(mol)$, $i = 0, \ldots, N - 1$. For the vapour rate, the value $V = 11(mol/h)$ was taken. The final time $t_f = 0.8(h)$ assures that the reboiler is never empty: indeed, it corresponds to 88% the time of the complete emptying of the reboiler with maximal distillate rate $u = V$. In addition, we took $x_N = 0.1$ and $y_1 = 0.95$. In order to obtain the distillate of desired purity, $N \geq N_{\text{min}}$ plates are needed. As it be seen from Tab.1 and Fig.2, the minimal number of plates depends on the location of the graph of the function $y(x)$ with respect to the diagonal, the separation is more difficult if this graph contains inflection points, as in the case of benzene - ethylendiamine.

3.2 Numerical solution by BOCOP solver

| mixture               | $N$ | $U_a(t_f)$ | recovery rate | discretization scheme |
|-----------------------|-----|------------|---------------|-----------------------|
| hexane - p-xylene     | 5   | 1.39534    | 93.35%        | Lobatto, 1600 points  |
| p-xylene              | 9   | 1.88885    | 98.58%        | Gauss, 1800 points    |
|                       | 12  | 2.20997    | 99.02%        | Gauss, 1800 points    |
|                       | 14  | 2.42212    | 99.21%        | Gauss, 1800 points    |
| methanol - water      | 6   | 1.3557     | 86.21%        | Lobatto, 1600 points  |
| benzene - ethylendiamine | 9  | 1.7188     | 92.24%        | Gauss, 1800 points    |
|                       | 12  | 2.1734     | 98.67%        | Gauss, 1800 points    |

Numerical results discussed below were obtained with the BOCOP optimal control solver (Bonnans et al. (2014)). BOCOP is an open-source toolbox for solving optimal control problems by direct method (Trelat (2005)). The optimal control problem is approximated by a finite dimensional optimization problem (NLP) resulting from the time discretization of the dynamics of the system by an appropriate choice of the discretization scheme. In the current version different options are available starting from the 1-st order explicit Euler scheme until 6-th order implicit Lobatto algorithm. The NLP problem is then solved by Ipopt algorithm.

For the numerical resolution the full formulation (5) of the problem was used. Tab.2 displays the optimal costs and the recovery rates of the light component corresponding all examples considered below. In general, the convergence of the algorithm is very sensitive to the chosen discretisation scheme, and it is particularly difficult when the number of plates is close to the minimal one. The most results presented below were obtained with the 4-th order implicit Gauss method with 1800 discretisation points.

Case 1: hexane - p-xylene The optimal control policies of the maximum distillate problem for this mixture are shown in Fig. 3 for different number of plates. The first column ($N = 5$) has just enough plates to obtain the distillate of desired quality. We observe that in this case the optimal control is almost of bang - bang type with very short singular arcs, there are 21 commutations between different types of controls. The first and the last control arcs correspond to the maximal distillate rate $u = V$. As we will see below, this is always the case in all studied examples. In total there are 10 maximal distillate rate intervals alternating with zero distillation rate intervals. Very short singular control arcs precede the maximal
Case hexane-p-xylene, $N = 5$: comparison of the switching function $H_1$ (thin red curve) and the rescaled optimal control $u/100$ (thick black curve) distillate rate arcs 2, 3 and 4. Such a control policy is very similar to the so-called cyclic operating policy described in Sorensen (1999).

BOCOP allows to extract from its output data the values of the adjoint vector $p$ along the optimal solution. This makes possible to perform the a-posteriori computation of the switching function $H_1(p,q)$. In Fig. 4 we compare the behaviour of this function with the optimal control $u(t)$ (taken with the rescaling factor 0.01). As expected, the sign-changes of $H_1$ are perfectly correlated with the switches between bang and singular arcs of the control function. So, the optimal control $u(t)$ computed numerically by direct method via complete discretization verifies the necessary optimality conditions of PMP.

With the second column ($N=9$) the separation is much easier. The corresponding optimal control has 4 bang arcs of maximal distillate rate, 2 arcs of zero control, and two singular arcs: the first one lasts about 25% of the total operation time $t_f$, and there are 8 switching points between different control types. In the case $N = 12$ the number of switchings drops to 5. The first bang arc (of maximal distillate rate) became longer, and there is only one short singular arc preceding the maximum distillate rate arc in the middle. Finally, with $N = 14$ the optimal control is of bang-bang type with one maximal distillate rate arc in the middle, which separates two long zero control arcs.

Case 2: methanol-water

The optimal distillate rate policy of this mixture is shown in Fig. 5. For the first configuration, $N = N_{\text{min}} + 2 = 6$ we found a bang-bang type control policy. The control switches 16 times between 9 maximal distillate rate arcs and 8 zero control arcs. Again, at the beginning and at the end the maximal control policy is used. The increment of the number of plates makes easier the separation. The optimal control policy becomes more tricky, but the number of switchings drops up to 9. Three singular arcs appear in the case $N = 9$. The first short singular arc connects two maximal rate arcs, the second connects the first zero control arc with the third maximal rate arc, and the third arc, of very duration, precedes the terminal bang arc. The duration of the second singular arc is about 43% of $t_f$. With the further increment of $N$ ($N = 12$) the first and the third singular arcs becomes longer, while the second shortens.

Case 3: benzene-ethylenediamine

Fig. 6 gives the idea of the optimal distillate rate policy for benzene-ethylenediamine mixture. For $N = 9$ plates we observe the picture similar to the previous two examples, though quality of the numerical solution is much worth compared to the

Globally, we observe the same tendency as in the previous example, which make us think that with the bigger number of plates the optimal control policy would be of bang-bang type with very few switching points.
previous ones. Again, we observe a quasi bang-bang structure, which disappears with the increment of the number of plates. In the last picture ($N = 12$) the appearing singular arcs alternates with the maximal and zero distillate rate arcs. The series starts and terminates with two bang arcs $u = V$.

The above examples put in evidence the dependence of the optimal solution of the maximum distillate problem with the separation task difficulty and the column configuration. If the number of plates is close to the minimal one, the optimal control is almost of the bang-bang type with a number of short maximum rate arcs. With bigger number of plates the same quality of the final product can be achieved with smaller number of commutations by combining bang and singular type controls. If the number of plates in the columns is big enough, the operation can be done by a simple series of bang controls. In any case, the amount of the final product increases with the number of plates, as well as the recovery arc of the light component. Such a mechanism cannot be (and was not) identified if the dynamics of the internal plates of the column is considered in a steady-state approximation, as was done in the literature so far.

### 3.3 Validation in dynamical simulation

The maximum distillate problem formulated in Section 2 is based on a series of quite strong assumption on the real dynamics of the column: equimolar overflow, total condensation, independence of the thermodynamic properties of the mixture on temperature. In order to test if the optimal solution obtained for the simplified problem can bring a real improvement to the conventional operation techniques, a more realistic simulation was done using the ProSim software (ProSim (2000)). We considered a 5 plates column charged with hexane and p-xylene, modeled by a UNIFAC equations. A bang-bang scenario approximating the optimal solution presented on the top of Fig. 3 was proposed. The column was driven to a steady-state and then the bang-bang scenario was realized. In Tab.3 the results of this simulation are compared with the solution obtained by BOCOP and with the solution obtained by ProSim with conventional operation policy. With almost the same control policy, the recovery rate in the "real" column is smaller than the one obtained by BOCOP used in the column configuration. In the "real" column is smaller than the one obtained by ProSim with conventional operation policy. With almost the same control policy, the recovery rate obtained by ProSim with conventional operation policy. The solution obtained by BOCOP and with the solution obtained by BOCOP, the optimal controls can be of bang or singular types, which correspond to three types of reflux policy: infinite reflux, zero reflux and singular reflux that can be computed from PMP. We presented the results of the numerical resolution of the problem by direct method using the BOCOP optimal control solver. Our results put in evidence the strong correlation between the structure of the optimal distillate policy and the number of the plates in the column, in relation to the thermodynamics of the binary mixture to be separated. We show that a cyclic-like policy reported by other authors in a different context is very close the optimal solution of the problem if the number of plates is just sufficient or is big enough to assure the desired purity of the distillate. If the number of plates is relatively small, but far from $N_{min}$, the optimal way to obtain the product of the desired quality consist in the alternation of the singular controls with the periods of zero or maximal distillate rate policies.

### Table 3.

| $x_d$ | ProSim (bang-bang) | Prosim (conventional control) |
|-------|---------------------|------------------------------|
| 95.33% | 96%                 | 97%                          |
| recovery rate | 93.35%       | 86%                          | 76%                          |

### 4. CONCLUSION

This paper focuses on the batch distillation of homogeneous binary mixtures. We discuss the correct optimal control formulation of the maximum distillate problem using the distillate rate instead of reflux as the control parameter. By introducing a new state variable, the purity deviation $z$, we reduce the distillate purity constrain to a simple terminal condition for $z$. According to PMP, the optimal controls can be of bang or singular types, which correspond to three types of reflux policy: infinite reflux, zero reflux and singular reflux that can be computed from PMP. We presented the results of the numerical resolution of the problem by direct method using the BOCOP optimal control solver. Our results put in evidence the strong correlation between the structure of the optimal distillate policy and the number of the plates in the column, in relation to the thermodynamics of the binary mixture to be separated. We show that a cyclic-like policy reported by other authors in a different context is very close the optimal solution of the problem if the number of plates is just sufficient or is big enough to assure the desired purity of the distillate. If the number of plates is relatively small, but far from $N_{min}$, the optimal way to obtain the product of the desired quality consist in the alternation of the singular controls with the periods of zero or maximal distillate rate policies.

### REFERENCES

F. Bonnans, D. Giorgi, V. Grélard, S. Maindrault, P. Martinon. BOCOP: User Guide. www.bocop.org.
B. Bonnard, M. Chyba. Singular Trajectories and their Role in Control Theory. Springer Science & Business Media, Vol. 40, 2003
A.O. Converse, G.D. Gross. Optimal Distillate-Rate Policy in Batch Distillation. Ind. Eng. Chem. Fundamen., 2 (3), pp. 217 – 221, 1963.
U.M. Diwekar. Unified approach to solving optimal design-control problems in batch distillation. AIChE, 38 (10), pp. 1551 – 1563, 1992
M.F. Doherty, M.D. Malone. Conceptual Design of Distillation Systems. MCGraw Hill chemical engineering series, 1st ed., 568 p., 2001.
S. Farhat, L. Pibouleau, S. Domenech. Optimal control of batch distillation via nonlinear programming. Chemical Engineering Process, 29, pp.33 – 38, 1991.
K.J. Kim, U.M. Diwekar. New era in batch distillation: computer aided analysis optimal design and control. Reviews in Chemical Engineering, 17(2), pp. 111 – 164, 2001.
I.M. Mujtaba. Batch distillation design and operation. Imperial college Press, 2004.
www.prosim.net
E. Sorensen A cyclic operating policy for batch distillation Theory and practice. Computers and Chemical Engineering, 23 (4-5), pp. 533-542, 1999.
E. Trelat E. Contrôle optimal : Théorie & applications. Vuibert, Collection "Mathématiques Concrètes", Ist ed. 246 p. 2005
S.R. Upreti. Optimal Control for Chemical Engineers. CRC PRES, 2011.