Optimization of Multiple-Fraction Batch Distillation by Nonlinear Programming

Batch distillation processes are very attractive for the recent development of the chemical industry: multipurpose, flexible plants and fine chemistry. For many separations of high-added value products, even a modest change in operating conditions has a significant economic impact—there is an important challenge for optimizing such processes.

Short-cut and dynamic models are the two classical approaches to the simulation of batch distillation columns. For problems without holdup, an intermediate procedure based on a decoupling method is validated.

For a multifraction separation problem with fixed final time, the reflux policies for each period and the period switching times constitute the set of decision variables. For predefined reflux policies, we apply an engineering approach to the solution of a such constrained variational problem, based on its transformation into a nonlinear programming problem. In this computer-implementable algorithm, the gain in distillate for the optimal linear or exponential reflux policies is significant (about 10%) compared with the optimal constant reflux policy.

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Introduction

In multipurpose, flexible plants, batch distillation constitutes an efficient separation tool consisting of a series of operation steps with different goals and specifications on product purities. This separation technique is used when a component is to be extracted from variable or complex charges, or if an impurity accumulation may occur during a continuous distillation task. Even though batch distillation consumes more energy than continuous distillation, it provides more flexibility and involves less capital investment (Luyben, 1988). A single column can also treat a wide range of feed compositions, a number of components, and degrees of difficulty of separation. The flexibility of this separation method makes it possible to adjust the operating conditions to the required purity specifications. The operation can be carried out with constant reflux ratio, with constant overhead compositions, or with an optimal policy suitable for the problem under consideration.

The mixture to be separated is all initially charged into the reboiler; during the first period, the light component product (main-cut or production-cut P) is collected into a product tank until the average composition drops a specified purity level and then the first intermediate distillate fraction (off-cut or slop-cut S) is produced during the second period. The procedure is repeated until the heaviest component reaches a desired purity in the column reboiler. This type of operation is also called multiple-fraction batch separation. Since for many separations of high-added value products even a modest change in time and/or in main cuts has a significant economic impact, there is a great incentive for improving the operation of such columns.

When multiple-fraction separations are considered, several objectives must be reached so that the conventional approaches based on dynamic programming or variational calculus lead to complex formulations. Indeed, in solving multidimensional problems, the limitations of dynamic programming due to its excessive demand in computer time and storage are well known. For variational calculus, the explicit solution of the Euler–Lagrange equations can be quite difficult and time-consuming; so from an engineering point of view, it is preferable to convert the variational problem to an approximating (due to predefined reflux policies), nonlinear programming (NLP) problem and solve it using existing NLP codes.

For a multiple-fraction separation problem with fixed final

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distillation time, the decision variables are the reflux policies for each period and the period switching times. One way to achieve their optimization is to repeatedly solve the problem of optimal reflux policies for fixed switching times and then locate an approximation of the optimal solution by iterating the procedure in the time-space. Obviously, such a method would demand excessive computer time.

After analyzing the most significant papers in the batch distillation optimization field, a simulation model is presented and validated with a classical example. For predefined shapes of reflux policies, the general optimal control problem of a multiple-fraction separation process is then formulated as an NLP problem, where the goal is to simultaneously optimize switching times and reflux policies. The choice of the reflux policy shape can be made in a wide range of functions such as constant, linear, polynomial or exponential and represents the single decision that the operator or the designer must make. Subsequently, this NLP approach is illustrated by three examples involving constant, linear and exponential/constant reflux policies; these examples show that the optimal linear or exponential reflux policies give significant gains of the amount of distillate, compared with the optimal constant reflux policy.

Previous Works

The optimization of a batch distillation column is generally considered in the literature as an optimal control policy problem. Especially the determination of optimal reflux policies has induced many researchers to solve one of the following well-known problems:

1. *Maximum distillate problem:* collection of maximum amount of distillate with specified concentration in a prescribed final time (Converse and Gross, 1963; Keith and Brunet, 1971; Murty et al., 1980; Diwekar et al., 1987)
2. *Minimum time problem:* minimization of final distillation time to produce a given amount of distillate with specified concentration (Coward, 1967a, b; Robinson, 1969, 1970; Mayur and Jackson, 1971; Egly et al., 1979, 1983; Hansen and Jörgensen, 1986; Mujtaba and Macchietto, 1988)
3. *Maximum distillate and minimum time problem:* collection of maximum amount of distillate with specified concentration in a minimum distillation time (Gangiah and Husain, 1974; Farhat et al., 1989)
4. *Maximum profit problem:* optimization of some economic criterion (Kerkhof and Vissers, 1978; Diwekar et al., 1989).

Problems 1 and 2 are usually solved by means of variational calculus or Pontryagin's maximum principle. Recently Mujtaba and Macchietto (1988) solved problem 2 with recycle of the off-cut for a binary mixture by using a successive quadratic programming (SQP) method.

The solution of problem 3 is obviously more complex and is based on the Pontryagin's maximum principle in the space control coupled with some specific algorithms [conjugate gradient method for Gangiah and Husain (1974) and dichotomy search for Farhat et al. (1989)].

Kerkhof and Vissers (1978) solved problem 4 by applying the Pontryagin's maximum principle, and Diwekar et al. (1989) used a generalization of the Davidon's method enabling linear constraints to be dealt with. In the latter case the multiple-fraction problem is treated as a number of single-fractions in a series.

A quite different approach (factor capacity methodology) is used by Luyben (1988) who gives several results on the effects of various design parameters for ternary mixtures.

In the greatest part of published works, the optimal solution of problem 1, 2, 3 or 4 is searched by using simplifying assumptions made either on thermodynamic models or on the holdup. Furthermore, the batches charged into the reboiler are generally binary mixtures or multicomponent mixtures considered as binary ones.

An ideal equilibrium for ternary mixtures is assumed by Mayur and Jackson (1971), and a linear equilibrium relationship is used by Keith and Brunet (1971).

The liquid holdup is either generally neglected (Converse and Gross, 1963; Coward, 1967a, b; Robinson, 1969; Keith and Brunet, 1971; Gangiah and Husain, 1974; Kerkhof and Vissers, 1978; Murty et al., 1980; Hansen and Jörgensen, 1986; Diwekar et al., 1987; Diwekar et al., 1989) or considered only as binary mixtures (Mayur and Jackson, 1971).

Multicomponent batch distillation is treated in a limited number of articles (Robinson, 1969, 1970; Mayur and Jackson, 1971; Egly et al., 1979, 1983; Diwekar et al., 1987; Mujtaba and Macchietto, 1988). These authors, however, have studied only the maximum recovery of a given cut or the multiple-fraction problem is treated as a succession of independent subproblems where the termination conditions for the previous fraction are the initial conditions for the following fraction (Diwekar et al., 1989).

The solution of the maximization of all the cuts by means of the maximum principle should lead to solving a multiple-criteria decision-making problem, because the optimal reflux policies for each cut depend on the switching times, which are themselves parameters to be optimized. So this approach appears to be very complex to implement.

Recently, Logsdon and Biegler (1989) suggested a collocation-based nonlinear programming formulation for obtaining optimal control profiles.

However, when the reflux policies are predefined for each period, the criterion becomes integrable and the problem can be formulated as an NLP problem, as will be discussed subsequently. From this new formulation, the maximum recovery of all the production cuts can be carried out for a fixed final time with specified purities. These purity constraints are computed from the simulation model mentioned earlier.

Simulation Model

Modeling equations

To treat multicomponent mixtures in columns involving high numbers of plates within moderate CPU times, the assumptions used by Domenech and Enjalbert (1981) have been retained:

- Theoretical plates
- Negligible liquid holdup
- Negligible pressure drop
- Total condenser
- Vapor and liquid which flow constantly into the column
- No feed stream and intermediate sidestream
- Thermodynamic constants which depend only on the temperature.

In the simulation model, used to optimize the column outputs (amount of cuts) under the purity constraints on distillate compositions, the heaviest component compositions at each step time and the condenser temperature are not included. So the process in Figure 1 can be described by the following set of

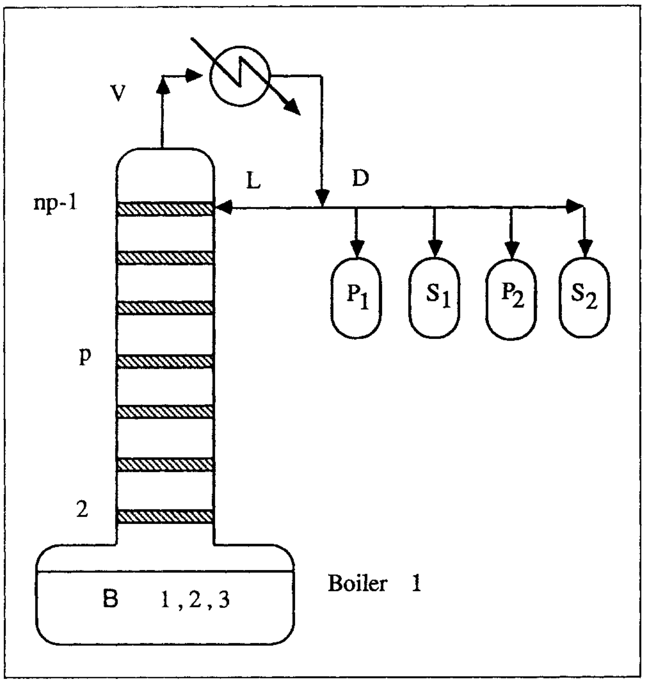


Figure 1. Batch distillation column for a ternary mixture.

differential and algebraic equations:

Overall mass balance

$$\frac{dB}{dt} = \frac{-V}{(R+1)} = -D \tag{1}$$

Mass balance on the (n − 1) lightest components between the condenser and the reboiler

$$B\frac{dX_{i,1}}{dt} = \frac{-V}{(R+1)}(X_{i,np} - X_{i,1}) \quad i = 1 \text{ to } n-1 \tag{2}$$

Summation equation on liquid compositions at the reboiler

$$1 - \sum_{i=1}^{n} X_{i,1} = 0 \tag{3}$$

Summation equation on vapor compositions at the reboiler

$$1 - \sum_{i=1}^{n} K_{i,1}(T_1)X_{i,1} = 0 \tag{4}$$

Partial mass balance on the (n − 1) lightest components between a plate p and the condenser

$$K_{i,p}(T_p)X_{i,p} - \frac{R}{(R+1)}X_{i,p+1} - \frac{1}{(R+1)}X_{i,np} = 0$$
$$i = 1 \text{ to } n-1; \quad p = 1 \text{ to } np-1 \tag{5}$$

Equilibrium equation on vapor compositions at plate p

$$1 - \sum_{i=1}^{n} K_{i,p}(T_p)X_{i,p} = 0 \quad p = 2 \text{ to } np-1 \tag{6a}$$

where $X_{i,np}$ is given by:

$$X_{i,np} = 1 - \sum_{i=1}^{n-1} X_{i,p}$$

So Eq. 6a becomes

$$1 - K_{n,p}(T_p) - \sum_{i=1}^{n-1}[K_{i,p}(T_p) - K_{n,p}(T_p)]X_{i,p} = 0$$
$$p = 2 \text{ to } np-1 \tag{6b}$$

Summation equation on liquid compositions at the condenser

$$1 - \sum_{i=1}^{n} X_{i,np} = 0 \tag{7}$$

Solution procedure

The set of differential and algebraic equations (DAE's) (Eqs. 1 to 7) is solved by a decoupling method from steady-state startup with total reflux.

The solution of the differential system (Eqs. 1 and 2) is obtained from a Runge–Kutta fourth-order procedure with variable step size and gives $(n-1)$ compositions into the reboiler. Equations 3 and 4 are solved separately to determine the heaviest component liquid composition and the temperature in the reboiler. These values are reported into the algebraic system (Eqs. 5 and 6), which is solved by an iterative Newton–Raphson procedure to obtain the $(n-1)$ distillate compositions. Then the summation equation (Eq. 7) gives the heaviest component composition. These compositions are likewise reported in the differential system (Eqs. 1 and 2) and the next integration step is carried out. The procedure is repeated until the final time is reached. However, the Fortran code of the model has been implemented so as to be called for a given period j; in this case, the initial conditions are the values of the state vector at the end of period $j-1$, and the outputs of the procedure are the amounts of distillate and the average composition for the key component for period j.

The DAE's (Eqs. 1 to 7) involve the following variables:

- Reboiler: number of moles (1), temperature (1), liquid compositions (n)
- Intermediate plates $p = 2$ to $np - 1$: temperatures $(np-2)$, liquid compositions of the $(n-1)$ lightest components $(np-2)(n-1)$
- Condenser: liquid compositions (n).

This DAE system can be decomposed as follows:

- n differential equations (Eqs. 1 and 2)
- $(n-1)(np-1) + (np-2)$ algebraic equations (Eqs. 5 and 6b)
- Three independent equations (Eqs. 3, 4 and 7).

Therefore, the model contains $n.np + 2$ variables and $n.np + 2$ equations; the flow chart of the solution procedure is presented in Figure 2.

The linearized equations (Eqs. 5 and 6b) are arranged plate by plate to consider at each calculation step two consecutive plates and the condenser. So the Jacobian matrix has a block-bidiagonal right-bordered structure as it is shown in Figure 3 for a four-component example with seven plates (the nonnull terms are represented by crosses and the null coefficients are not

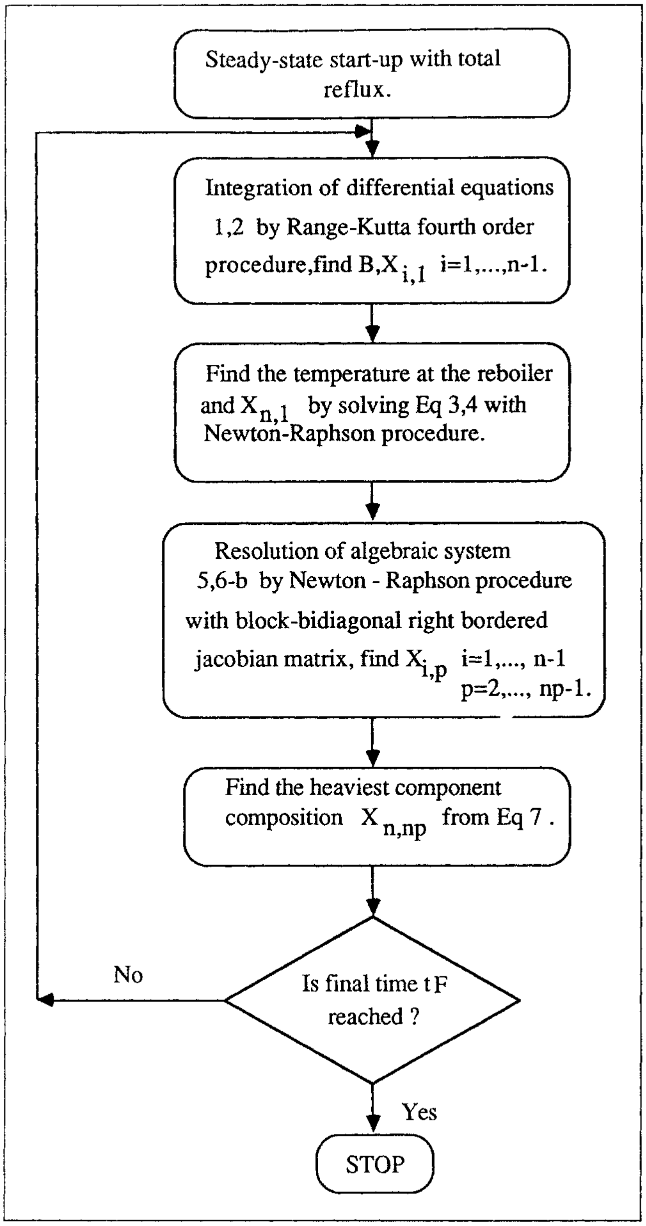


Figure 2. Flow chart of the simulation procedure.

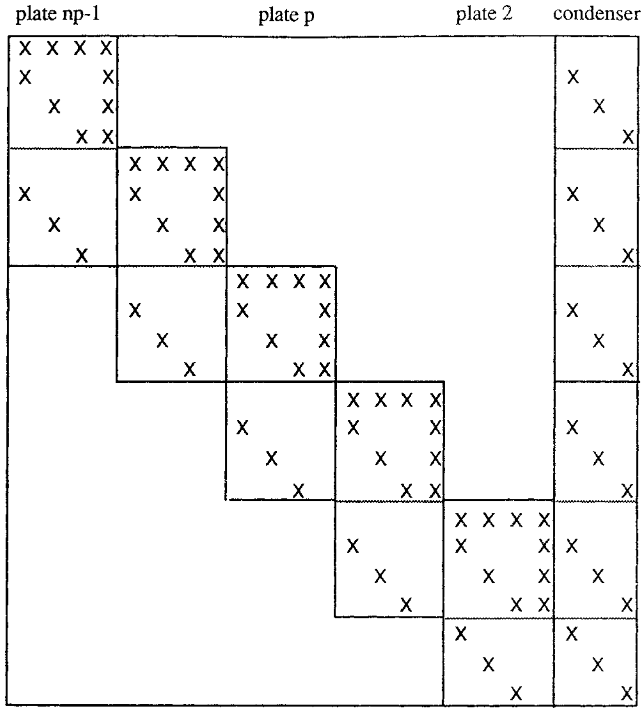


Figure 3. Structure of the Jacobian matrix of the linearized system (n = 4, np = 7).

reported). The first row in each block represents Eq. 6b, and the remaining $(n - 1)$ rows are Eq. 5 while the first $(n - 1)$ columns represents the lightest component compositions and the final column is the temperature. The solution of the nonlinear system is obtained from the classical Newton–Raphson procedure.

Simulation example

Let us consider the separation of feed stream containing ten components by means of a column operated with constant reflux ratio $R = 20$ and involving 22 plates and a total condenser. In this high-sized example, presented by Domenech and Enjalbert (1981), the overall holdup in the column represents only 3% of the batch charged into the reboiler, and so it can be neglected.

The data are reported in Table 1 and the changes with time of the distillate compositions for each component obtained by the model without holdup and the model presented by Domenech and Enjalbert (1981) are superimposed in Figure 4. For columns with low overall holdup a good agreement between the two models can be noted, thus allowing the model without holdup to accurately describe the batch rectification process within moderate CPU times—about one minute on a BULL DPX 5045 computer (equivalent to 5 Vax unit performance).

NLP Solution of the Batch Rectification Problem

The batch rectification process described by the DAE's system (Eqs. 1 to 7) can be represented by:

$$f[t, x(t), x^o(t), R(t), v] = 0, \quad t \in [t_0, t_F] \tag{8}$$

where t is the independent variable (time), $x(t) \in R^l$ is the vector of all state variables (number of moles in the reboiler, compositions and temperatures, $l = n.np + 2$), $x^o(t)$ represents

Table 1. Data of the Example by Domenech and Enjalbert

Comp. No.	Boiling Point, °C	Thermodynamic Coefficients A	B	C	Molec. Weight	Mol % in Feed
1	184.4	7.63846	1,976.3	231	93.1	0.6385
2	228.5	7.17331	1,779.1	186	127.6	8.2176
3	245.0	7.96718	2,502.2	247	215.9	0.5173
4	246.6	7.98568	2,550.2	253	162.0	3.0917
5	263.2	8.07435	2,680.9	253	162.0	41.5835
6	268.4	8.69275	3,146.6	273	250.3	0.8421
7	272.5	8.65385	3,149.1	273	162.0	0.5333
8	284.0	7.97685	2,654.8	237	286.0	28.3411
9	303.3	7.81129	2,520.8	208	303.0	1.5953
10	318.4	7.73620	2,638.2	225	316.0	14.6382

$\log K = A - B/(T + C)$

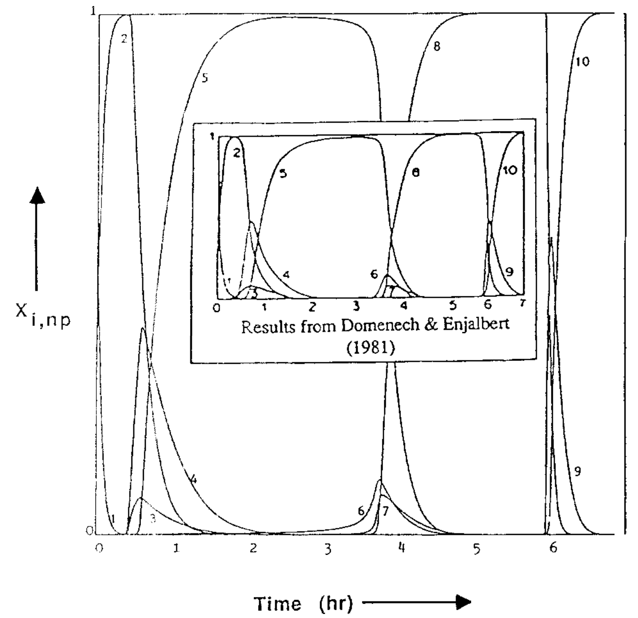
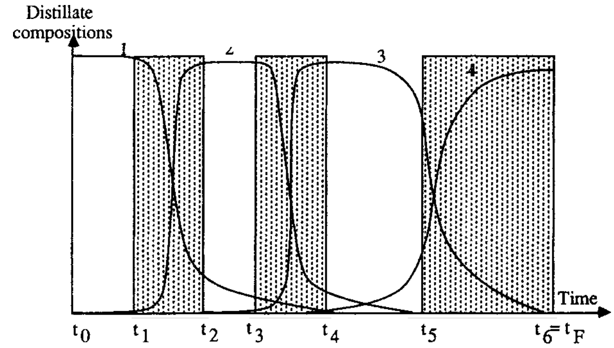


Figure 4. Results of Domenech and Enjalbert (1981) vs. this work.



Periods	1	2	3	4	5	6
Weighting factors	β_1	β_2	β_3	β_4	β_5	β_6
Cuts	P_1	S_1	P_2	S_2	P_3	S_3 (and P_4 at t_F)
Reflux policies	R_1	R_2	R_3	R_4	R_5	R_6
Key components	1	-	2	-		

Figure 5. Succession of production cuts and off-cuts for a quaternary mixture.

The last cut is collected into the reboiler.

the derivatives of $x(t)$ with respect to time, $R(t) \in R$ is the control variable (reflux ratio), and $v \in R^{n+2}$ is the vector of time invariant parameters ($n - 1$ initial compositions, operating pressure, number of plates, and vapor flow rate); to simplify the following developments, the vector v will not be mentioned. The time interval of interest is $[t_0, t_F]$ where t_0 is the initial time and t_F the given final time of the whole operation.

The multiple-fraction batch distillation process is a sequential system described by a series of periods (main cut or off cut), where the initial conditions for each period are the final state of the previous period. For a given period j, the formulation (Eq. 8) becomes:

$$f_j[t, x(t), x^o(t), R_j(t)] = 0, \quad t \in [t_{j-1}, t_j] \tag{9}$$

where t_{j-1} is the initial time of the period j, t_j is the final time of this period, and $R_j(t)$ is the reflux ratio function during this period. For a mixture involving n components, it exists $2n - 2$ alternate periods productions and off-cuts (see Figure 5), because the heaviest component is recovered into the reboiler; so the index j lies in the interval $[1, 2n - 2]$. If the index j is an odd number [i.e., the period $[t_{j-1}, t_j]$ is a production period for the key component number $k = (j + 1)/2$), the system is submitted to a purity terminal constraint on the kth key component, this equality constraint occurs when $t = t_j$:

$$g_k[t_j, x(t_j), x^o(t_j), R_j(t)] = 0 \tag{10}$$

The system performance is measured in terms of the following objective function to be maximized for the main cuts or minimized for the off-cuts:

$$F_j = \int_{t_{j-1}}^{t_j} \frac{V}{R_j(t) + 1} dt \tag{11}$$

where V is the vapor flow rate.

The sequential nature of the batch rectification problem leads to the following multiple criteria decision-making (MCDM) problem:

$$\max_{R_1(t), t_1} F_1$$

under constraint g_1

$$\min_{R_1(t), R_2(t), t_1, t_2} F_2$$

under constraint g_1

$$\max_{R_1(t), R_2(t), \ldots, R_j(t), t_1, t_2, \ldots, t_j} F_j \; (j \text{ odd}) \tag{12}$$

under constraints $g_1, g_2, \ldots, g_{(j+1)/2}$

$$\min_{R_1(t), R_2(t), \ldots, R_{j+1}(t), t_1, t_2, \ldots, t_{j+1}} F_{j+1}$$

under constraints $g_1, g_2, \ldots, g_{(j+1)/2}$

$$\max_{R_1(t), R_2(t), \ldots, R_{2n-2}(t), t_1, t_2, \ldots, t_{2n-2} = t_F} F_{2n-1}$$

under constraints $g_1, g_2, \ldots, g_n$.

At the junction points between the two periods, the purity constraints must be verified, that is why for example constraint g_1 is written for period 2 and so on.

The solution of the MCDM problem (Eq. 12) is always done by converting it to a scalar optimization problem:

$$\max_{\mathcal{R}, \mathcal{T}} \sum_{j=1}^{2n-1} \beta_j F_j \tag{13}$$

under constraints $\mathcal{G} = (g_1, g_2, \ldots, g_n)$ where $\mathcal{R}$ is the set of the reflux ratio $R_1(t), R_2(t), \ldots, R_{2n-2}(t)$, $\mathcal{T}$ is the set of switching times $t_1, t_2, \ldots, t_{2n-2}$, and β_j is a positive (respectively negative) factor for a main cut (or off-cut). In practical industrial applications, the weighting factors β_j can be assimilated to economic factors.

In a batch distillation process, the problem generally consists of maximizing the production of main cuts within a given cycle time so that a reduced form of Eq. 13 becomes

$$(P1)\ \underset{\mathcal{R},\mathcal{T}}{\text{Max}} \sum_j \beta_j F_j \quad j \in [1, 3, 5, \ldots, 2n-1] \tag{14}$$

under the set of constraints $\mathcal{G}$.

When the shape of the reflux policy $R_j(t)$ is chosen in such a way that the objective function F_j (Eq. 11) is integrable in each period j, Eq. 14 can be reformulated as an NLP problem. As it is shown in Figure 5, the functions F_j of the optimization problem (Eq. 14) are the production cuts P_k $[k = (j+1)/2]$.

Another solution of minimizing the weighted sum of off-cuts is

$$(P2)\ \underset{\mathcal{R},\mathcal{T}}{\text{Min}} \sum_j \beta_j F_j \quad j \in [2, 4, 6, \ldots, 2n-2] \tag{15}$$

under the set of constraints $\mathcal{G}$. where the functions F_j are the off-cuts S_k $(k = j/2)$. In problem $(P2)$, the weighting factors β_j either have an economic significance (recycling or marginal costs) or a technical interpretation (storage requirements).

Formulation of problem (P1)

An example of formulation of problem (P1), when the reflux policies are assumed to be linear during each production period, is discussed in this section. Other examples of formulations with different reflux shapes (constant and exponential) are given in the Appendix. By isolating the last production cut, the problem (P1) can be written as follows:

$$(P1)\ \underset{\mathcal{A},\mathcal{B},\mathcal{T}}{\text{Max}} \left\{\beta_{2n-1} P_n + \sum_j \beta_j P_k\right\}$$

$$j \in [1, 3, 5, \ldots, 2n-3]; \quad k = (j+1)/2 \tag{16}$$

$$P_k = \int_{t_{j-1}}^{t_j} V\,dt/(R_j + 1) = \int_{t_{j-1}}^{t_j} V\,dt/(a_j t + b_j + 1) \tag{17}$$

where k is the index of the key component which is collected at the end of period j and

$$\mathcal{A} = a_1, a_2, \ldots, a_{2n-2}$$
$$\mathcal{B} = b_1, b_2, \ldots, b_{2n-2}$$
$$\mathcal{T} = t_1, t_2, \ldots, t_{2n-3}.$$

The last production term has been isolated from the general summation, because it is expressed from the overall mass balance:

$$P_n = B_0 - \sum_{i=1}^{n-1} (P_i + S_i) \tag{18}$$

When $a_j \neq 0$, the integration of Eq. 17 is obvious and problem $(P1)$ is the following:

$$\underset{\mathcal{A},\mathcal{B},\mathcal{T}}{\text{Max}} \left\{\beta_{2n-1} B_0 + \sum_j [(\beta_j - \beta_{2n-1}) V/a_j \ln [(a_j t_j + b_j + 1)/(a_j t_{j-1} + b_j + 1)] - \beta_{2n-1} V/a_{j+1} \ln [(a_{j+1} t_{j+1} + b_{j+1} + 1)/(a_{j+1} t_j + b_{j+1} + 1)]]\right\}$$

$$j \in [1, 3, 5, \ldots, 2n-3]; \quad k = (j+1)/2 \tag{19a}$$

with the nonlinear purity constraints (computed from the simulation model):

$$g_k = \overline{X}_{k,np} - \overline{XSPEC}_k$$

that is to say

$$g_k = \frac{\int_{t_{2k-2}}^{t_{2k-1}} \frac{V X_{k,np}(t)}{R_{2k-1} + 1}\,dt}{\int_{t_{2k-2}}^{t_{2k-1}} \frac{V}{R_{2k-1} + 1}\,dt} - \overline{XSPEC}_k = 0 \tag{19b}$$

$$g_n = \overline{X}_{n,1} - \overline{XSPEC}_n = 0 \tag{19c}$$

where $\overline{X}_{n,1} = X_{n,1}(t_F)$.

The linear constraints are related to the switching times

$$t_j \leq t_{j+1} \quad j = 1, 2, \ldots, 2n-3 \tag{19d}$$

Furthermore, all the variables are physically bounded.

Formulation of problem (P2)

When the reflux policies are assumed to be linear, the problem $(P2)$ can be expressed as follows:

$$(P2)\ \underset{\mathcal{A},\mathcal{B},\mathcal{T}}{\text{Min}} \left\{\sum_j \beta_j S_k\right\}$$

$$j \in [2, 4, 6, \ldots, 2n-2]; \quad k = j/2 \tag{20}$$

$$S_k = \int_{t_{j-1}}^{t_j} V\,dt/(R_j + 1) = \int_{t_{j-1}}^{t_j} V\,dt/(a_j t + b_j + 1) \tag{21}$$

When $a_j \neq 0$, the problem $(P2)$ becomes:

$$(P2)\ \underset{\mathcal{A},\mathcal{B},\mathcal{T}}{\text{Min}} \left\{\sum_j \beta_j V/a_j \ln [(a_j t_j + b_j + 1)/(a_j t_{j-1} + b_j + 1)]\right\}$$

$$j \in [2, 4, 6, \ldots, 2n-2] \tag{22}$$

under the set of constraints (Eqs. 19b, 19c and 19d) ($\mathcal{A}$, $\mathcal{B}$, $\mathcal{T}$ are the same vectors as in the previous case).

The problems, $(P1)$ and $(P2)$, have the same dimension, that is to say the same numbers of optimization variables, linear constraints, and nonlinear ones. Table 2 presents the variation of this dimension vs. the number of feed components.

Table 2. **Dimension of Problems ($P1$) and ($P2$) for Linear Reflux Policies**

No. of Comp.	2	3	4	5	10
Periods	2	4	6	8	18
Decision Variables	5	11	17	23	53
Linear Constraints	1	2	3	4	9
Nonlinear Constraints	2	3	4	5	10

Numerical Examples

In the first two following examples, the NLP problem is solved by means of an augmented Lagrangian procedure described by Pibouleau et al. (1985). We consider the ternary system involving components 1, 2 and 7 in Table 1; and Table 3 presents the input data for these two examples.

In the last example, the problem ($P2$) is solved by using the same numerical procedure, for exponential reflux policies during the production cuts and constant policies during the off-cuts. This example on the separation of three hydrocarbons into a 20-plate column was simulated by Nad and Spiegel (1987). We assumed a constant vapor molar flow of 2,690 mol · h^{-1} and the mixture ideality; the other data are reported in Table 4.

The three problems involve five operational steps P_1, S_1, P_2, S_2, and P_3 and must be solved for fixed final times t_F.

Example 1: solution of (P1) for constant and linear policies

The problem ($P1$) is solved for $\beta_1 = 1, \beta_3 = 1$, and $\beta_5 = 0$: the main cuts P_1 and P_2 have similar importance and the last cut P_3, which is collected into the reboiler, is an uninteresting product. So the purity of P_3 is not specified, and consequently the rectification stops at the end of the production cut P_2 ($t_F = t_3$). The problem is to maximize $P_1 + P_2$ for predefined constant and linear reflux policies. In both cases, the specifications for components 1 and 2 are 95%, and the final time t_F is equal to 2.3 hours.

Due to the implicit nature of constraints g_k, their convexity cannot be proved; so, the optimization problem may be nonconvex or may have several local optima. However, because the results were obtained from several starting points, one can think that the problem is at least locally convex. Table 5 shows that the optimal linear reflux policy offers 11.1% more distillate than the optimal constant reflux policy. The distillate compositions and optimal reflux policies are shown in Figures 6a and 6b. Discontinuities observed on the distillate composition curves are due to the assumption of instantaneous switching at a period boundary. In a more accurate model, this switching would be simulated by a valve cut-off.

Example 2: solution of (P2) for constant and linear policies

The problem ($P2$) is solved for $\beta_2 = 1$ and $\beta_4 = 1$, thus minimizing the sum $S_1 + S_2$, which is equivalent to maximizing the sum of production cuts $P_1 + P_2 + P_3$, according to the overall mass balance, $S_1 + S_2 = B_0 - (P_1 + P_2 + P_3)$. When all the weighting factors β_i are identical, the solution of ($P2$) is then equivalent to the solution of ($P1$).

The problem is solved for constant and linear reflux policies, in which the specifications are 95% for components 1 and 3, and 92.5% for intermediate component—the final time is fixed at 2.5 hours. As in the previous example, due to the lack of information about the problem convexity, the results presented in Table 6 were obtained from several starting points. A significant gain of 10.7% can be observed between the optimal linear reflux policy and the optimal constant reflux policy. Figures 7a and 7b show the distillate compositions and the two optimal policies.

Example 3: solution of (P2) for constant and exponential/constant policies

As in the previous case, the problem ($P2$) is solved for $\beta_2 = 1$ and $\beta_4 = 1$. For practical purposes, exponential reflux policies, $R_j(t) = A_j \exp(\omega_j t)$, are implemented during the production cuts, and constant policies, $R_{j+1}(t) = R_{j+1}$, are chosen for the off-cuts; this approach is commonly used in industrial batch separation processes. For final time t_F fixed at 6.25 hours and given specifications of 85.5% for cyclohexane, 91% for *n*-heptane and 92% for toluene, the NLP procedure gives the optimal

Table 3. **Input Data for Examples 1 and 2**

$V = 110$ mol · h^{-1}	$B_0 = 100$ mol
$X_{1,1} = 0.33$	$P = 761.654$ torr
$X_{2,1} = 0.33$	(Pa = torr × 133)
$X_{3,1} = 0.34$	$np = 7$

Table 4. **Input Data for Example 3**

$B_0 = 2{,}930$ mol	$P = 761.654$ torr
$V = 2{,}690$ mol · h^{-1}	(Pa = torr × 133)
$X_{1,1} = 0.407$	$np = 20$
$X_{2,1} = 0.394$	1:Cyclohexane
$X_{3,1} = 0.199$	2:*n*-Heptane
	3:Toluene

Component	Thermodynamic Coefficients A	B	C
Cyclohexane	6.85146	1,206.407	223.136
n-Heptane	6.89386	1,264.370	216.640
Toluene	6.95087	1,342.310	219.187

$\log K = A - B/(T + C)$

Table 5. **Optimal Solution of Problem ($P1$) for Constant and Linear Reflux Policies**

Cuts	Amount of Cuts Collected: R Constant	R Linear
P_1	30.29	32.10
S_1	5.86	2.80
P_2	22.34	26.37
$P_1 + P_2$	52.63	58.47

Optimal Values of R Constant

t_1	t_2	R_1	R_2	R_3
1.091	1.544	2.963	7.506	2.720

Optimal Values of R Linear

t_1	t_2	a_1	b_1	a_2	b_2	a_3	b_3
1.095	1.354	3.878	1.021	6.151	1.668	3.293	−2.868

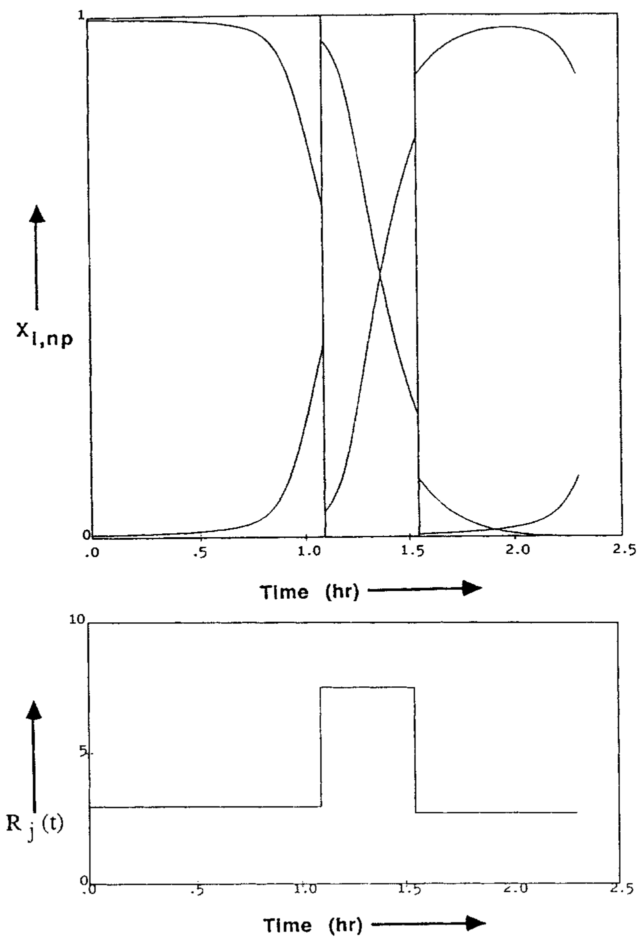


Figure 6a. Distillate compositions obtained by solving (*P*1): optimal constant policy.

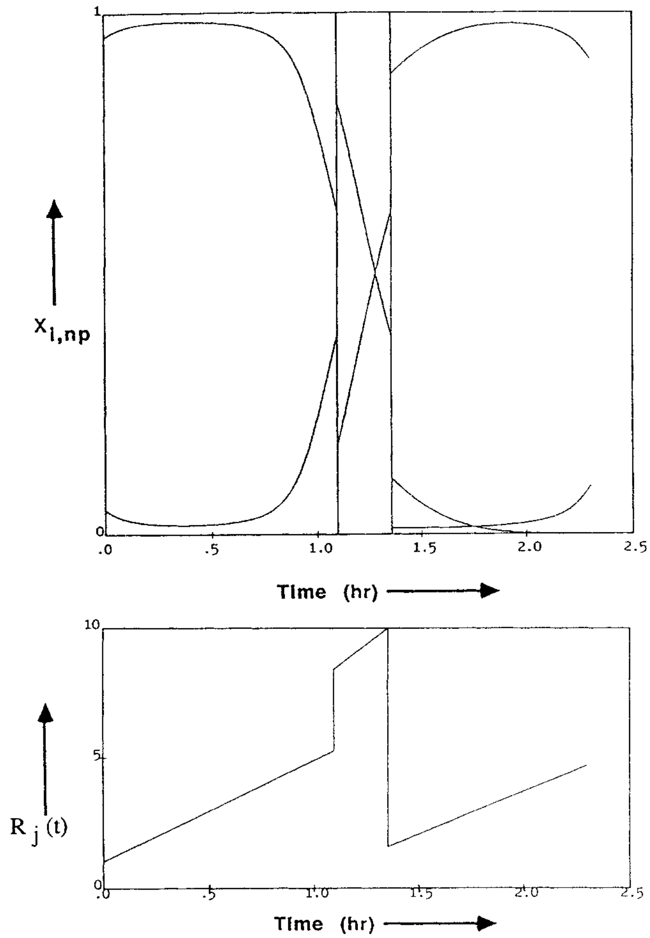


Figure 6b. Distillate compositions obtained by solving (*P*1): optimal linear reflux policy.

values of terms A_j, ω_j and R_{j+1} for each production cut j and off-cut $j + 1$. The same example is also solved with constant reflux policies for each cut, as shown in Table 7; and the variation in the duration of distillate compositions and reflux are shown in Figures 8a (constant policies) and 8b (exponential/constant policies). A significant gain of 5.5% on the total production is obtained with the exponential/constant policy, as shown in Table 7.

Table 6. Optimal Solution of Problem (*P*2) for Constant and Linear Reflux Policies

Cuts	Amount of Cuts Collected: R Constant	Amount of Cuts Collected: R Linear
P_1	29.48	31.23
S_1	6.31	3.68
P_2	25.31	29.62
S_2	7.55	0.98
P_3	31.35	34.49
$P_1 + P_2 + P_3$	86.14	95.34

Optimal Values of R Constant

t_1	t_2	t_3	R_1	R_2	R_3	R_4
0.983	1.347	2.231	2.669	5.345	2.838	2.925

Optimal Values of R Linear

t_1	t_2	t_3	a_1	b_1	a_2	b_2	a_3	b_3	a_4	b_4
0.973	1.229	2.345	3.981	0.848	5.233	0.918	3.224	−2.359	7.341	−1.507

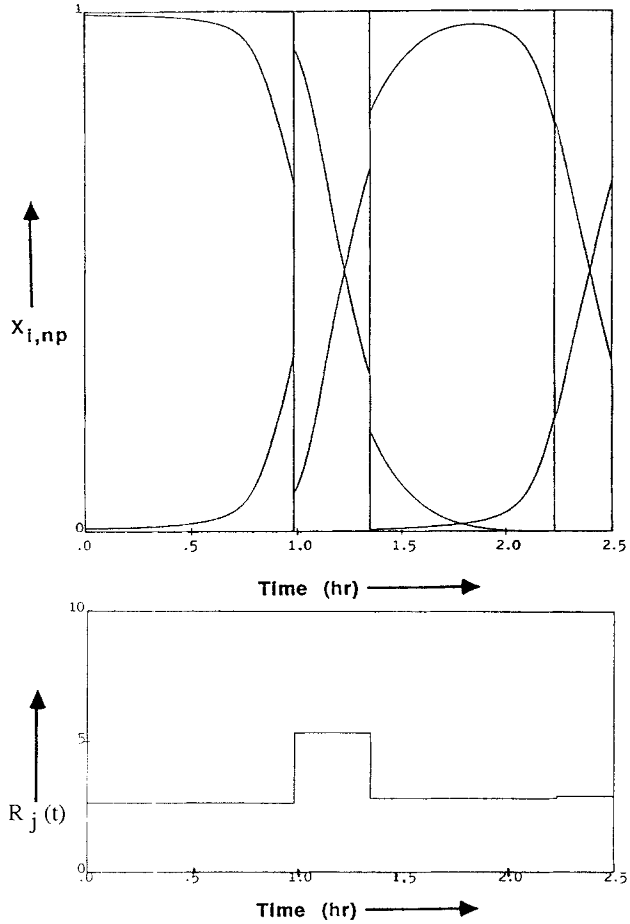


Figure 7a. Distillate compositions obtained by solving (*P*2): optimal constant policy.

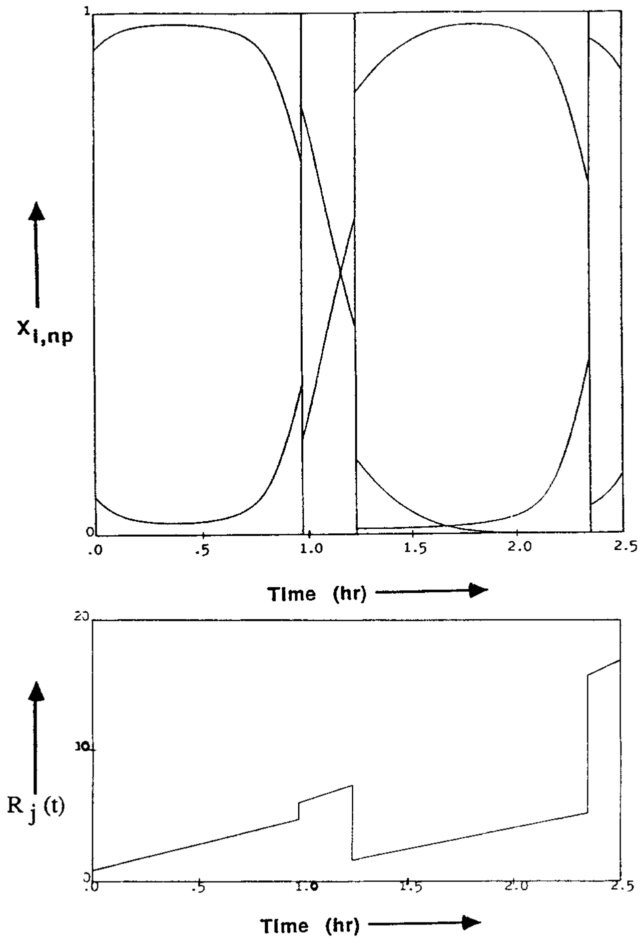


Figure 7b. Distillate compositions obtained by solving (*P*2): optimal linear reflux policy.

Conclusions

A new approach for solving the modeling equations of a batch distillation process without holdup is presented. The decoupling procedure for solving the DAE system is validated using a published example. Its low CPU time and its ability to accurately describe the physical problem enable the procedure to be used for optimization.

The optimization of the multiple-fraction separation problem

Table 7. Optimal Solution of Problem (*P*2) for Constant and Exponential/Constant Reflux Policies

Cuts	Amount of Cuts Collected: *R* Constant	Amount of Cuts Collected: *R* Exponential/Constant
P_1	1,151.1	1,182.5
S_1	493.1	264.3
P_2	673.3	825.3
S_2	30.1	125.8
P_3	582.4	532.1
$P_1 + P_2 + P_3$	2,406.8	2,539.9

Optimal Values of R Constant

t_1	t_2	t_3	R_1	R_2	R_3	R_4
2.038	3.288	6.038	3.760	5.816	9.985	17.950

Optimal Values of R Exponential/Constant

t_1	t_2	t_3	A_1	ω_1	R_1	A_2	ω_2	R_2
2.017	3.274	6.031	1.888	0.680	11.789	0.049	1.163	3.701

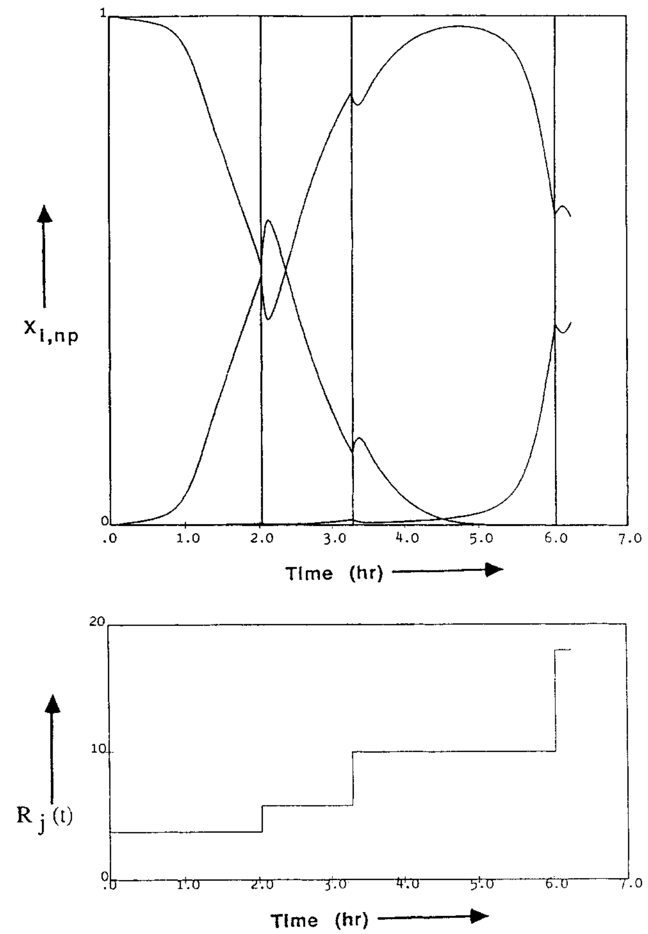


Figure 8a. Distillate compositions obtained by solving (*P*2): optimal constant policy.

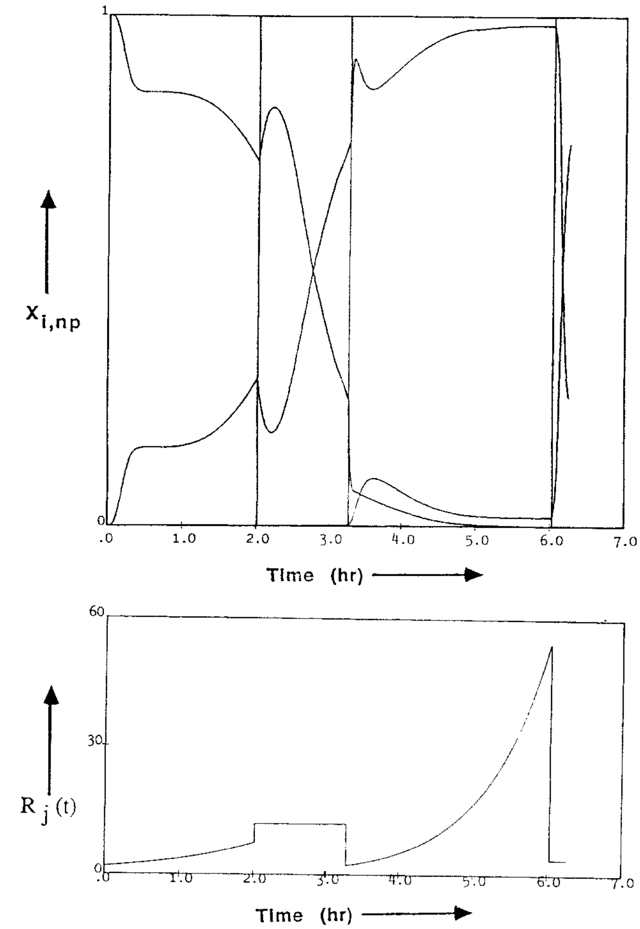


Figure 8b. Distillate compositions obtained by solving (*P*2): optimal exponental/constant reflux policy.

with fixed final time is also developed. This is accomplished by simultaneously optimizing the switching times and the reflux policies for each period. From an industrial point of view, it is often desirable to maximize the productions for a fixed final time. Indeed, the batch distillation process generally forms part of a complex plant, and the determination of processing times of a sequence of unit operations is given by solving a scheduling problem.

By assuming predefined reflux policies, the multiple-criteria decision-making problem, whose direct solution is practically impossible, is reformulated as an NLP problem. The reflux policies are chosen as constant, linear, or exponential in each period (production and off-cut). The constant reflux policy is easily implementable on an industrial batch column, and the linear or exponential reflux trajectories can in practice be followed by a set of control increments. The NLP problem can be solved either by maximizing the weighted sum of production cuts [problem (*P*1)] or by minimizing the weighted sum of off-cuts [problem (*P*2)], which are equivalent problems when all the weighting factors are the same. Three numerical examples have shown that the optimal linear or exponential reflux policies give about 5 to 10% more distillate compared with the optimal constant reflux policy.

According to the problem dimension, the initial values and the shape of the predefined reflux policy, the mean CPU time required for reaching the optimal solution is about two hours on a BULL DPX 5045 computer.

Notation

- $\mathcal{A}$ = set of slopes for linear reflux ratio
- A_j = constant coefficient for exponential reflux for period j
- a_j = slope of linear reflux ratio for period j
- $\mathcal{B}$ = set of intercept times for linear reflux ratio
- B = amount of moles in the reboiler, mol
- B_0 = initial amount of moles in the reboiler, mol
- b_j = intercept time for linear reflux ratio for period j
- D = distillate flow rate, mol · h^{-1}
- F_j = objective function for period j
- f_j = system of differential-algebraic equations for period j
- $\mathcal{G}$ = set of purity constraints
- g_k = purity constraint for kth key component
- $K_{i,p}$ = equilibrium coefficient for component i on plate p
- n = number of components
- np = number of plates in the column (including reboiler and condenser)
- P = pressure, torr
- P_k = production cut related to the kth key component, mol
- $\mathcal{R}$ = set of predefined reflux policies
- R = reflux ratio
- R_j = reflux ratio for period j
- S_k = off-cut following P_k, mol

T_p = temperature on plate p, °C
$\mathcal{T}$ = vector of switching times
t = time, h
t_0 = initial time, h
t_F = final time, h
t_j = switching time for period j
V = vapor flow-rate, mol · h^{-1}
$X_{i,p}$ = liquid composition of component i on plate p
$\overline{X}_{k,np}$ = average composition of component k collected in the tank
$XSPEC_k$ = specified average composition for component k collected

Greek letters

β_j = weighting factor for period j
ω_j = constant coefficient for exponential reflux for period j

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Appendix: Problem Formulation for Reflux Shapes

The maximum production-cut formulation [problem ($P1$)] is:

$$\text{Max}\left\{\beta_{2n-1}P_n + \sum_j \beta_j P_k\right\}$$
$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A1)}$$

where

$$P_n = B_0 - \sum_{i=1}^{n-1}(P_i + S_i)$$
$$= B_0 - \sum_j (P_k + S_k) \quad j = 1, 3, 5, \ldots, 2n-3; k = \frac{(j+1)}{2}$$
$$= B_0 - \sum_j\left[\int_{t_{j-1}}^{t_j}\frac{V}{(R_j(t)+1)}dt + \int_{t_j}^{t_{j+1}}\frac{V}{(R_{j+1}(t)+1)}dt\right]$$
$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A2)}$$

Equation A1 becomes:

$$\text{Max}\left[\beta_{2n-1}B_0 + \sum_j\left\{(\beta_j - \beta_{2n-1})\int_{t_{j-1}}^{t_j}\frac{V}{(R_j(t)+1)}dt - \beta_{2n-1}\int_{t_j}^{t_{j+1}}\frac{V}{(R_{j+1}(t)+1)}dt\right\}\right]$$
$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A3)}$$

Constant reflux policy

Let $R_j(t) = R_j$ and $R_{j+1}(t) = R_{j+1}$, where R_j and R_{j+1} are constant reflux to be optimized for production and off-cut periods; so the production quantities at the head of the column are integrable:

$$P_k = \int_{t_{j-1}}^{t_j}\frac{V}{(R_j+1)}dt = \frac{V(t_j - t_{j-1})}{(R_j+1)}$$
$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A4)}$$

and

$$S_k = \int_{t_j}^{t_{j+1}}\frac{V}{(R_{j+1}+1)}dt = \frac{V(t_{j+1} - t_j)}{(R_{j+1}+1)}$$
$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A5)}$$

For all periods $j = 1, 3, 5, \ldots, 2n-3$, Eq. A1 becomes:

$$\text{Max}\left[\beta_{2n-1}B_0 + \sum_j\left\{(\beta_j - \beta_{2n-1})\frac{V(t_j - t_{j-1})}{(R_j+1)} - \beta_{2n-1}\frac{V(t_{j+1} - t_j)}{(R_{j+1}+1)}\right\}\right]$$
$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A6)}$$

which is a classical NLP form for problem ($P1$).

Exponential/constant reflux policy

The reflux policy is now assumed to be an exponential function for the production period $R_j(t) = A_j \exp(\omega_j t)$ and constant for the off-cut period $R_{j+1}(t) = R_{j+1}$. Therefore, the optimization variables are A_j, ω_j, and R_{j+1}. The integration of the overhead production cut P_k leads to:

$$P_k = \int_{t_{j-1}}^{t_j} \frac{V}{(A_j \exp(\omega_j t) + 1)} dt$$

$$= V\left[(t_j - t_{j-1}) - \frac{1}{\omega_j} ln \frac{(1 + A_j \exp(\omega_j t_j))}{(1 + A_j \exp(\omega_j t_{j-1}))}\right]$$

$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A7)}$$

and the integral of S_k is given by Eq. A5; so the NLP form for problem $(P1)$ is:

$$\text{Max}\left[\beta_{2n-1} B_0 + \sum_j \left\{(\beta_j - \beta_{2n-1}) V\left[(t_j - t_{j-1}) - \frac{1}{\omega_j} ln \frac{(1 + A_j \exp(\omega_j t_j))}{1 + A_j \exp(\omega_j t_{j-1})}\right] - \beta_{2n-1} \frac{V(t_{j+1} - t_j)}{(R_{j+1} + 1)}\right\}\right]$$

$$j = 1, 3, 5, \ldots, 2n-3; \quad k = (j+1)/2 \quad \text{(A8)}$$

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