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The Time-Optimal Problem in Binary Batch Distillation with a Recycled Waste-Cut

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ABSTRACT

Recently Pontryagin's maximum principle has been applied to find the time-optimal reflux policy to achieve a given separation of two components by batch distillation. In the case where a sequence of identical charges is available, it is possible to improve upon this optimal policy by exploiting the extra degrees of freedom that result if a waste-cut is permitted. This is demonstrated with the aid of a specific example, using both piecewise constant and piecewise continuous reflux policies. Two methods of solution are discussed.

INTRODUCTION

The purpose of a binary batch distillation is to separate a charge of specified quantity and composition into a distillate and a residue, each with specified composition. Two well-known methods of accomplishing this are the so-called 'constant reflux' and 'constant overheads' modes of operation. In the former, the reflux ratio is held constant throughout the distillation, at that value which ensures that the accumulated distillate and the residue simultaneously attain the specified compositions. The second method, on the other hand, seeks to maintain the instantaneous distillate composition at its desired value at all times, by continually increasing the reflux ratio during distillation.

Clearly, these two policies do not exhaust all the possibilities. Indeed, the freedom to choose other reflux policies was used by Coward[1] to minimise the total time required for a specified separation, while Converse and Gross,[2] using the method of dynamic programming, solved the equivalent problem of maximising the amount of distillate of a specified composition obtainable in a given time.

Neglecting hold-up in the column, Coward's work provides a complete solution to the problem of separating a given charge into a given distillate and a given residue in minimum time. However, batch distillation is often used to separate a quantity of material much larger than the capacity of the kettle, by distilling a sequence of charges, and in this case, modes of operation more general than those considered by Coward are available. In particular, it is no longer necessary that each charge be separated into a distillate and a residue simultaneously meeting the imposed specifications, since any off-specification material may be returned to the kettle, as a part of the next charge, for reprocessing. This suggests the possibility of periodic operation, in which each charge consists of fresh feedstock mixed with the recycled off-specification material from the previous distillation. Each batch separation, then, consists of two parts; in the first, a quantity of accumulated overheads meeting the distillate specification is collected; in the second, the residue is further distilled until it meets the bottoms composition specification. The overhead produced during the second part meets neither specification, and is referred to as the 'waste-cut'. It is recycled as part of the charge for the next batch.

With this kind of operation, it is necessary to consider the choice of reflux ratio during each part of the distillation, and a variety of problems results from imposing various constraints on these reflux ratios. The simplest is that of choosing a single constant reflux ratio throughout (i.e., the same in both parts). This is closely analogous to the 'constant reflux' method in simple batch distillation and is discussed below. Subsequently, we consider policies which do not require the reflux ratio to be the same for both parts of the operation; in other words, we admit piecewise-constant reflux policies. Finally, we eliminate the constraint of constancy, allowing continuous variation of the reflux ratio throughout each part of

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the separation, and thus solve a generalization of Coward's problem.

CONSTANT REFLUX POLICY

The batch still is assumed to consist of a kettle which can be charged initially with a mass B_c of feedstock of composition X_{Bc}. (Throughout this paper the term 'composition' is used to denote the mass fraction of more volatile component in the mixture.) The kettle is surmounted by a fractionating column, with performance corresponding to a given number of theoretical plates and in which the liquid and vapor hold-ups are negligible. The relative volatility (α) of the two components of the mixture is assumed constant, and the usual simplifying assumptions of constant liquid and vapor flow rates in the column are made. Upward vapor flow, which is equal to the boiling-rate in the kettle, is denoted by V, and is assumed constant throughout the distillation. Vapor leaving the top of the column is condensed, part is returned as reflux, and the remainder passes to a distillate accumulator. At any instant during distillation, the mass and composition of accumulated distillate are denoted by D and X_D, and the corresponding mass and composition of the residue in the kettle by B and X_B. When hold-up is negligible in the fractionating column and the condensing system, we have:

$$B_c X_{Bc} = DX_D + BX_B \tag{1}$$

and

$$B_c = D + B. \tag{2}$$

These equations hold throughout the distillation. The flow rate of distillate to the accumulator is dD/dt, and that of reflux down the column is $R\,dD/dt$, where R is the reflux ratio. Equating the sum of these to the flow of vapor leaving the column:

$$(R+1)\frac{dD}{dt} = V \tag{3}$$

or, in view of eqn. (2),

$$-(R+1)\frac{dB}{dt} = V \tag{3'}$$

The notation introduced here is illustrated in Fig. 1.

The well-known case of a simple batch distillation with constant reflux ratio is considered first. The charge is to be separated completely into a distillate of specified composition $\overline{X}_D$ and a residue of specified composition $\overline{X}_B$. Final quantities of distillate and residue are determined through the overall balance, eqns. (1) and (2), and the reflux ratio necessary to achieve the separation, which is uniquely determined by the specification of $\overline{X}_D$ and $\overline{X}_B$, can be found from the Rayleigh equation:

$$\ln\left(\frac{B_c}{\overline{B}}\right) = \int_{\overline{X}_B}^{X_{B_c}} \frac{dX_B}{Y_N(X_B, R) - X_B} \tag{4}$$

in which Y_N, a function of X_B and R, denotes the composition of vapor leaving the top of the fractionating column, which can be computed using plate to plate calculations through the column. A value of R satisfying eqn. (4) can be found providing the number of plates in the column is sufficiently large. At the beginning of the operation, the composition of accumulated distillate is better than specification and falls toward $\overline{X}_D$ as the distillation continues. At the same time, the concentration of the heavier component in the residue rises, and if the value of R is correctly chosen, the composition of the residue reaches the specified value $\overline{X}_B$ at the same time as the accumulated distillate attains the composition $\overline{X}_D$. The separation is then accomplished.

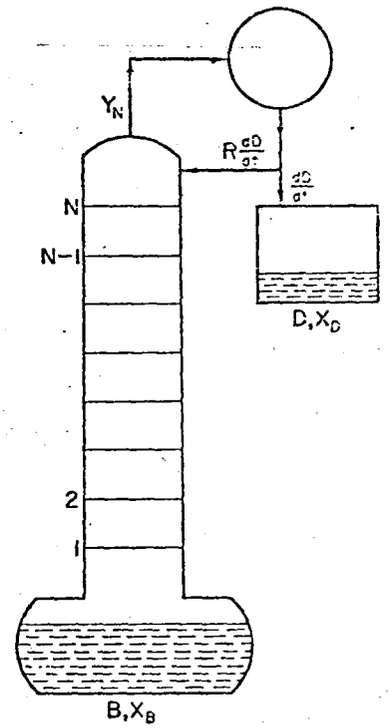


Fig. 1. Column and notation.

If a somewhat smaller value of R is chosen, the situation is similar, but when X_D falls to $\overline{X}_D$, X_B is still higher than $\overline{X}_B$. The accumulated distillate then meets its specification and may be taken as a product, but further distillation is necessary to bring the residue composition to $\overline{X}_B$. In the course of this, a second

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batch of distillate, which meets neither of the product specifications, accumulates as a 'waste-cut'. Since the reflux ratio is lower than that determined from eqn. (4), the total distillation time might be expected to be shorter, but a complete separation of the charge is not accomplished. In situations where the batch still processes a sequence of charges from the same feedstock, however, it is possible to recycle the waste-cut as part of the next charge to the kettle, and in this way one might hope to establish a cyclic operation in which the charge to the kettle is a mixture of fresh feedstock and the waste-cut from the previous distillation cycle. Such a mode of operation separates the feedstock into the desired products, and should require less time for distillation of each charge than the conventional constant-reflux separation. However, the quantity of fresh feedstock separated in each distillation is smaller than with the conventional method, since the kettle must accommodate the recycled waste-cut as well as fresh feed, and it is not immediately obvious whether the time *per unit of fresh feed distilled* is less than that obtained with the conventional method. This question is investigated in the present section.

If B_o is the amount of fresh feedstock in the charge, and X_{Bo} its composition, the course of the cyclic operation is as indicated in Fig. 2. The feedstock is

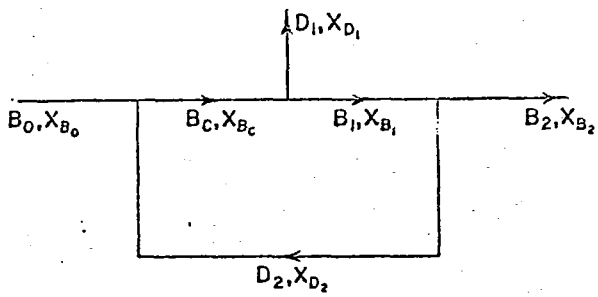


Fig. 2. Cyclic separation process.

mixed with a quantity D_2 of the waste-cut, of composition X_{D2}, to form the kettle charge B_c, of composition X_{Bc}. The distillation then proceeds until the composition X_{D1} of the accumulated distillate D_1 becomes equal to $\bar{X}_D$, when the distilled product is withdrawn. There remains a quantity B_1 of residue, of composition X_{B1}, in the kettle, and the distillation is continued until the quantity and composition of the residue become B_2 and X_{B2} respectively, where $X_{B2} = \bar{X}_D$. The accumulated distillate then forms the waste-cut, and is mixed with the next batch of fresh feed as indicated.

This cyclic state can be computed in various ways; for example, an initial calculation can be performed with $D_2^{(0)} = 0$, so that $[B_c^{(0)}, X_{Bc}^{(0)}] = [B_o, X_{Bo}]$. The resulting waste-cut $[D_2^{(1)}, X_{D2}^{(1)}]$ is then used in the second calculation, for which the kettle charge is formed by mixing the fresh feed $[B_o, X_{Bo}]$, with the waste-cut $[D_2^{(1)}, X_{D2}^{(1)}]$. The waste-cut $[D_2^{(2)}, X_{D2}^{(2)}]$ from this distillation is then used in forming the kettle charge for the third calculation, and so on, until successive calculations coincide to the desired accuracy. This is closely analogous to the physical procedure by which the column could be brought to the cyclic state; indeed, it differs only in that B_o is held constant through successive calculations, rather than the total kettle charge B_c. Calculations in which B_o is adjusted at each iteration to keep the value of B_c constant would also be perfectly practicable, and would be the exact analog of the physical start-up procedure.

Each stage in this iterative calculation was performed by integrating the pair of equations:

$$\frac{dX_B}{dt} = -\left[\frac{Y_N(X_B, R) - X_B}{B}\right]\left(\frac{V}{R+1}\right) \quad (5)$$

$$\frac{dB}{dt} = -\frac{V}{R+1} \quad (6)$$

from initial conditions B_c, X_{Bc}. Equation (5) is a differential form of the Rayleigh equation, and eqn. (6) a trivial rearrangement of eqn. (3'). The total distillation time was determined as the value of t at which $X_B = \bar{X}_B$. The values of B_1 and X_{B1} were found by locating the point on the solution of eqns. (5) and (6) at which the balance eqns. (1) and (2), were satisfied, with $X_D = \bar{X}_D$. The values of D_2 and X_{D2} follow from the balance equations for the second part of the distillation, namely:

$$B_1 X_{B1} = D_2 X_{D2} + B_2 X_{B2} \quad (7)$$

$$B_1 = D_2 + B_2 \quad (8)$$

Calculations of this type were carried out for a range of values of R below R_{max}, where R_{max} denotes the reflux ratio required to separate a charge of fresh feedstock completely into the specified products. This range is bounded below by the reflux ratio R_{min}, at which the first drop of distillate has composition $\bar{X}_D$, so that vanishingly small quantities of products are obtained, and the waste-cut composition approaches the feedstock composition. Data for the system were chosen to correspond to a case worked by Coward,[1] and are given in Table 1, while Fig. 3 shows the results of the computations, as distillation time T per 100 units fresh feedstock. The curve shows a clear minimum at a reflux ratio well below R_{max}, and it is seen that the distillation time in this case

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can be reduced by about 10%, compared with the conventional constant reflux policy.

TABLE I
PROBLEM SPECIFICATIONS

Number of stages	= 5
Vapor flow rate, V	= 143·10
Relative volatility	= 2
Fresh charge, B_0	= 100·00
Composition of fresh charge, X_{B_0}	= 0·40
Distillate specification, $\bar{X}_D$	= 0·85
Bottoms specification, $\bar{X}_B$	= 0·10

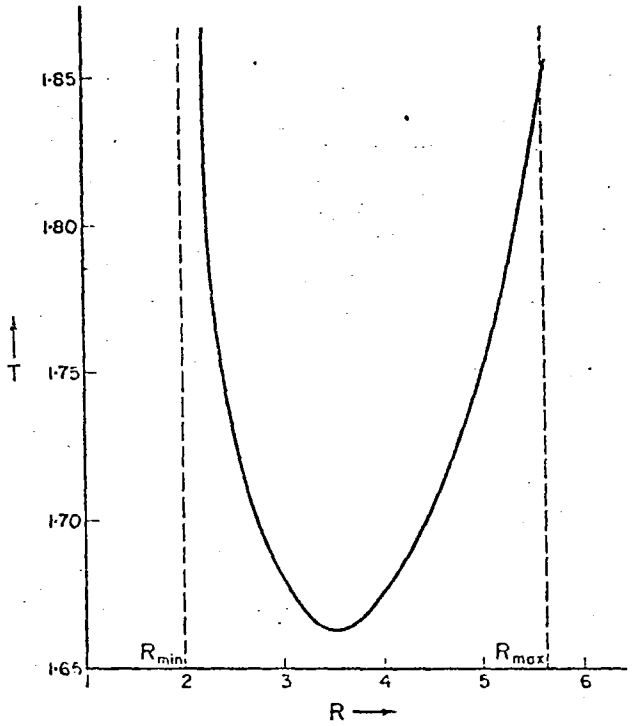


Fig. 3. Distillation time as a function of reflux ratio for the constant reflux policy.

PIECEWISE-CONSTANT REFLUX POLICY

The simplest generalisation of the above operating policy is obtained by admitting different constant values of the reflux ratio in the two parts of the distillation. These are denoted by R_1 and R_2, representing the reflux ratios before and after the first distillate cut is removed. The computational procedure described above can also be used in this slightly more general case; it is only necessary to use different values R_1 and R_2 of the reflux ratio in integrating eqns. (5) and (6) for the first and second stages of distillation, respectively. This procedure has two drawbacks: first, it converges only slowly in many cases; and secondly, there exist cases in which it never converges to a solution in which identical conditions are repeated in successive distillation cycles. Instead it sometimes approaches a state in which conditions repeat every second cycle, or occasionally, every third cycle. The former difficulty exists even in the calculations described in the last section, though now the slowness of the convergence is more serious, as many more calculations must be performed to explore behavior over the (R_1, R_2)-plane. The latter difficulty is serious, and makes it necessary to explore alternative computational schemes.

Fortunately, relaxation of the constraint that the reflux ratio should be the same in both stages permits a different and much faster computational scheme to be used. Referring to Fig. 2, if values of B_1 and X_{B1} are specified, overall mass balances determine all the quantities and compositions indicated. The reflux ratio R_1 required to achieve the separation of the charge (B_c, X_{B_c}) into the distillate (D_1, X_{D_1}) and the residue (B_1, X_{B_1}), is found from the integral form, eqn. (4), of the Rayleigh equation, which gives:

$$\ln\left(\frac{B_c}{B_1}\right) = \int_{X_{B_1}}^{X_{B_c}} \frac{dX_B}{Y_N(X_B, R_1) - X_B} \qquad (9)$$

Similarly, R_2 can be found from:

$$\ln\left(\frac{B_1}{B_2}\right) = \int_{X_{B_2}}^{X_{B_1}} \frac{dX_B}{Y_N(X_B, R_2) - X_B} \qquad (10)$$

The total distillation time then follows from eqn. (3′), which gives:

$$T = \left(\frac{R_1 + 1}{V}\right)(B_c - B_1) + \left(\frac{R_2 + 1}{V}\right)(B_1 - B_2) \qquad (11)$$

By choosing a number of pairs of values for B_1 and X_{B1}, the corresponding points in the (R_1, R_2)-plane and their associated distillation times can be calculated, and the results of such calculations are presented as a contour map of the function $T(R_1, R_2)$ in Fig. 4. The minimum is shallow and lies quite near to the diagonal $R_1 = R_2$, though not actually on it. Indeed, the optimal piecewise constant reflux policy has $R_1 = 3{\cdot}431$, $R_2 = 3{\cdot}485$ and gives a value of T only marginally less than the smallest obtained previously with a constant reflux ratio.

In the search for the optimum it must be realised that values of R_1 and R_2 cannot be found for all possible pairs of values of B_1 and X_{B_1}; it is clear, for example, that if $B_1 > B_c$ or $X_{B_1} > X_{B_c}$, no solution is possible. After performing one or two calculations with practicable values of B_1 and X_{B_1}, however, it becomes clear how they should be varied to obtain reasonable coverage of the interesting part of the (R_1, R_2)-plane.

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OPTIMUM REFLUX POLICY

The generalisation of Coward's optimum reflux problem to the present case is obtained by relaxing all constraints on the reflux ratio, except that it should be a piecewise-continuous function of time. The determination of the optimum reflux policy $R(t)$ in this class can be reduced to two problems of the

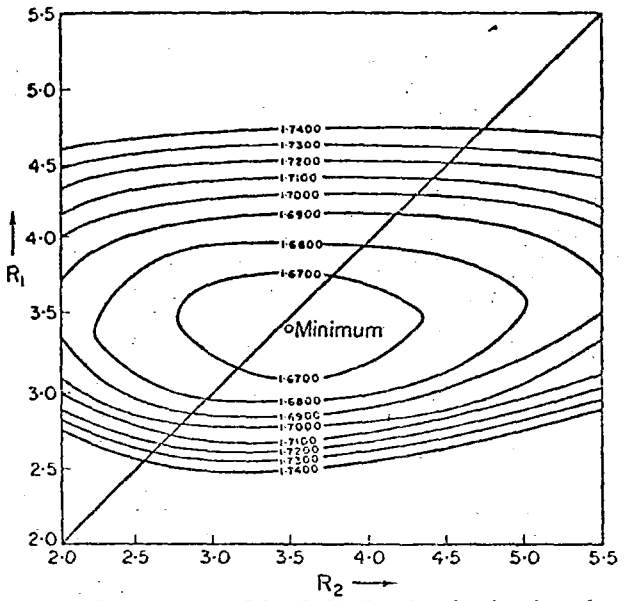


Fig. 4. Contour map of the distillation time for the piecewise-constant reflux policy.

type solved by Coward, using a procedure analogous to that introduced in the previous section. Suppose the optimum reflux policy consists of a function $R_1(t)$ during the first stage of distillation, when the charge (B_c, X_{Bc}) is separated into a distillate (D_1, X_{D1}), and a residue (B_1, X_{B1}), followed by a function $R_2(t)$ during the second stage of distillation, when the residue (B_1, X_{B1}) is separated into a waste-cut (D_2, X_{D2}) and a product (B_2, X_{B2}). Then, clearly, it is necessary that $R_1(t)$ should be such as to minimise the time required to pass from the state (B_c, X_{Bc}) to the state (B_1, X_{B1}), while $R_2(t)$ should be such as to minimise the time required to pass from (B_1, X_{B1}) to (B_2, X_{B2}). However, these conditions are not sufficient for optimality, since the values of B_1 and X_{B1} are not determined by the data, and it becomes necessary to search in the (B_1, X_{B1})-plane to determine the point at which the total distillation time is minimised.

For specified values of B_1 and X_{B1}, the determinations of both R_1 and R_2 are separately problems of the type discussed by Coward. Referring specifically to the first stage of distillation, we have:

$$\frac{dB}{dt} = -\frac{V}{R_1 + 1}; \quad B(0) = B_c, \quad B(T_1) = B_1 \tag{12}$$

$$\frac{dX_B}{dt} = -\left[\frac{Y_N(X_B, R_1) - X_B}{B}\right]\left(\frac{V}{R_1 + 1}\right); \quad X_B(0) = X_{B_c}, \quad X_B(T_1) = X_{B_1} \tag{13}$$

We introduce the variables p_1 and p_2, adjoint to B and X_B, respectively, satisfying the differential equations:

$$\frac{dp_1}{dt} = -p_2\left(\frac{Y_N - X_B}{B^2}\right)\left(\frac{V}{R_1 + 1}\right) \tag{14}$$

$$\frac{dp_2}{dt} = p_2\left(\frac{\frac{\partial Y_N}{\partial X_B} - 1}{B}\right)\left(\frac{V}{R_1 + 1}\right) \tag{15}$$

The Hamiltonian function is defined by:

$$H = -p_1\left(\frac{V}{R_1 + 1}\right) - p_2\left[\frac{Y_N(X_B, R_1) - X_B}{B}\right]\left(\frac{V}{R_1 + 1}\right) \tag{16}$$

and, according to the maximum principle of Pontryagin,[3] it is necessary for the optimality of $R_1(t)$ that:

(i) R_1 should be chosen to maximise H (with constant B, X_B, p_1, and p_2) for each $0 < t \leq T_1$, and
(ii) The (constant) maximum value of H should be positive.

Equations (12) through (15) can be integrated forward in time after guessing values of $p_1(0)$ and $p_2(0)$, and the guessed values must then be adjusted until $B(T_1)$ and $X_B(T_1)$ take their specified values B_1 and X_{B1}, and condition (ii) above is satisfied. Since eqns. (14), (15) and (16) are homogeneous in p_1 and p_2, only the ratio of the adjoints affects the values of the physical variables, so either of these variables may be assigned an arbitrary value at one time. For example, if the value of $p_1(0)$ is assigned, integration is continued until $X_B(T_1)$ falls to the value X_{B1}, and $p_2(0)$ is then adjusted until B simultaneously attains the value B_1. The corresponding time T_1 then represents the minimum time for the first stage of distillation. The minimum time T_2 for the second stage is determined in the same way, and the sum $T_1 + T_2$ gives the minimum distillation time subject to the constraint that (B, X_B) should take the values (B_1, X_{B1}) at the end of the first stage. The optimum reflux policy is then obtained by seeking the values of B_1 and X_{B1} which minimise this sum, and here this was accomplished by the sequential simplex search procedure of Spendley *et al.*,[4] in the (B_1, X_{B1})-plane.

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The search was started from the values of B_1 and X_{B1} found for the optimum piecewise-constant reflux policy in earlier calculations, and the final reflux policy is shown in Fig. 5.

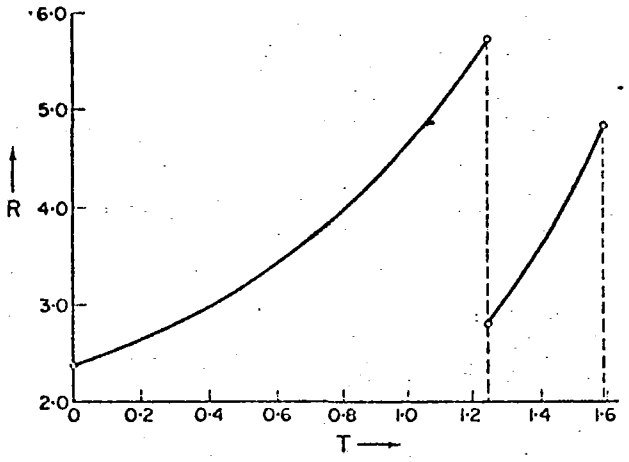


Fig. 5. Optimum piecewise-continuous reflux policy.

DISCUSSION

In Fig. 6 the conventional constant reflux policy, the conventional constant overheads policy and Coward's optimum policy for separation without a waste-cut, are compared with the optimum constant reflux policy, the optimum piecewise constant policy and the optimum piecewise continuous policy for separation with a waste-cut. The curves and their corresponding distillation times are identified in the caption to Fig. 6. It should particularly be noticed that the simplest constant reflux policy with a waste-cut gives a performance almost equal to that of the optimum reflux policy without a waste-cut, while change to the optimal piecewise continuous reflux policy yields a further improvement of about 3½%.

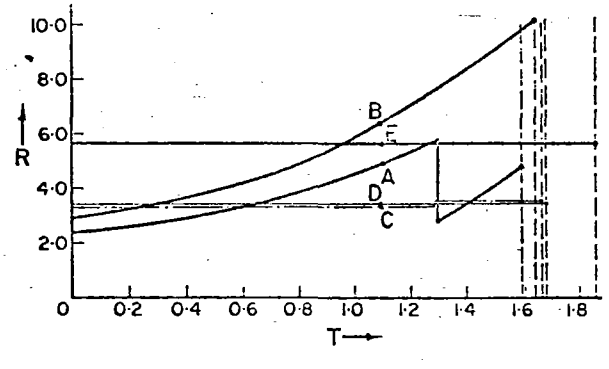


Fig. 6. Comparison of different reflux policies:

	Policy	*Curve no.*	*Distillation time*
Without waste-cut	Constant reflux	E	1·8526
	Constant overheads	–	2·0000
	Time optimal (Coward)	B	1·6450
With waste-cut	Constant reflux	D	1·6633
	Piecewise-constant reflux	C	1·6632
	Piecewise-continuous reflux	A	1·5973

Finally, it must be pointed out that the example used in the calculations was not chosen so that optimisation yielded great performance improvements. An easy separation which could be accomplished in a small number of plates was considered, so as to minimise the computational time, while illustrating the principles involved. Comparison with earlier work was facilitated by choosing specifications previously used by Coward.

ACKNOWLEDGEMENT

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NOMENCLATURE

B	mass of material in the kettle at any time
B_o, B_c	mass of fresh feed and that of the total charge to the still
B_1, B_2	masses of residue after the first and second stages of distillation
D	mass of accumulated distillate at any time
D_1, D_2	masses of distillate collected at the end of the first and second stages
H	the Hamiltonian function (defined in eqn. (16))
p_1, p_2	variables adjoint to B and X_B (defined in eqns. (14) and (15))
R_1, R_2	reflux policies in the first and second stages, respectively
T_1, T_2	time required for the first and second stages of distillation
T	total time for distillation (*i.e.* for one complete cycle)
V	constant boil up rate or vapor flow rate up the column, mass/time
X_B	mass fraction of the more volatile component in B (composition of B)
X_{Bo}, X_{Bc}	compositions of B_o and B_c, respectively
X_{B1}, X_{B2}	compositions of B_1 and B_2, respectively

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X_D composition of the distillate D at any time

X_{D1}, X_{D2} compositions of D_1 and D_2, respectively.

$\bar{X}_D, \bar{X}_B$ specified compositions of distillate and residue, respectively

Y_N composition of vapor from the top plate in the column

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RÉSUMÉ

On a appliqué récemment la méthode de Pontryagin à la recherche de l'évolution optimale du taux de reflux afin de séparer deux corps par distillation discontinue. Quand on dispose d'une séquence de charges identiques, on peut améliorer cette prévision optimale en exploitant les degrés de liberté supplémentaires dont on dispose si on peut se permettre du gaspillage... On démontre ceci à l'aide d'un exemple dans lequel on utilise d'une part un programme de reflux en escalier, d'autre part un programme d'évolution continue. On discute deux méthodes de solution.

ZUSAMMENFASSUNG

Vor kurzem wurde Pontryagin's Maximumprincip angewendet, um din zeitoptimalen Rückflussbetrieb zu finden und eine vorbestimate Trennung zweier Komponenten durch diskontinuierliche Destillation zu erreichen. Ist eine Folge gleicher Chargen verfügbar, so ist es möglich, das Endergebnis aufgrund dieser Optimalemethode zu verbessern, indem man die zusätzlichen Freiheitsgrade ausnützt, die aus der Zulassung eines 'waste-cut' resultieren. Dies wird anhand eines besonderen Beispiels unter Verwendung sowohl stückweise konstanter als auch stückweise kontinuierliche Rückflussbetriebe dargelegt. Zwei Lösungsmethoden werden besprochen.

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