ELSEVIER

Chemical Engineering Journal 71 (1998) 111–126

Chemical Engineering Journal

Optimal batch distillation control based on specific measures

B.H.L. Betlem*, H.C. Krijnsen, H. Huijnen

Department of Chemical Engineering, University of Twente, P.O. Box 217, 7500, AE Enschede, Netherlands

Received 17 February 1998; received in revised form 9 July 1998; accepted 16 July 1998

Abstract

Two independent measures characterise a single batch distillation run: the degree of separation difficulty, which indicates the difficulty at the start and the degree of exhaustion, which indicates bottom exhaustion at the end of the run. If one of both measures remains within bounds, then constant quality control appears to be the best control policy. It has been proven in the literature that the application of slop recycling increases the production rate. One of the goals of this study is to derive simple scheduling models based on specific measures for the optimal operation of batch distillation with slop recycling. Simulation studies for a cyclic pseudo-steady state operation over a broad range of degrees of difficulty for binary and ternary distillations are performed. Also the influence of the tray hold-up has been studied. All simulations show that at maximum production rate the degree of separation difficulty balances the degree of exhaustion (dependent variable in the optimum). The slop recycling strategy keeps the degree of exhaustion during the production phase and the degree of difficulty during the slop phase within bounds. As a result, the improvement of slop recycling at constant quality control compared to constant reflux control is 15–20% for difficult separation and can amount to more than 35% for relatively easy separations. The resulting production time appears to have a linear relation with the average separation difficulty. This relation has been experimentally verified. © 1998 Elsevier Science S.A. All rights reserved.

Keywords: Batch distillation; Simulation; Optimal control; Optimisation; Recycle policies

---

1. Batch distillation and operation objectives

A classical batch distillation process is considered. The feedstock is introduced into the bottom and distilled in a rectifier column producing the products overhead one by one. Consequently, during distillation not only the quality requirement of the current running product is of interest, but also the exhaustion of the remainder.

As indicated in Fig. 1, the general batch distillation run consists of a filling, a start-up, a number of sequential products and slop distillation cuts and a finishing phase. If the required bottom exhaustion cannot be realised during the product distillation, then slop distillate with a lower purity is produced until the required exhaustion is reached. This slop can be added to the feed of the next batch run. But also in cases in which, actually, slop recycling is not required, it still offers an opportunity for optimisation. Slop can improve the mean production rate, as bottom exhaustion during the production run takes relatively longer time.

Different slop recycle strategies are conceivable. Quintero-Marmol and Luyben [1] discuss six alternative forms of multi-component distillation. The objective is to maximise a profit function taking a fixed recharging time into account. The results show that mixing of different slop cuts with each other, slop cuts with feed amounts, or gradual feed addition during fed-batch distillation have a detrimental effect on the production rate. It is more profitable to take advantage of the separation already obtained.

In this article the slops will be retained separately: separate slop recycle. After each product phase a slop cut with a lower purity is produced. This cut is stored separately. Fig. 1 illustrates the connections between the sequential batch runs. The slop should be added to the next batch just before the production run of the most volatile component in the slop. All lighter components have been removed in the previous production run. By keeping all slops separately, already distilled liquid is not mixed, and in the long run less distillation effort will be required. Also, mixing-up will result in more interactions between the sequential runs. However, the former method will require extra storage capacity, which is a disadvantage. An economic evaluation has to make clear which method is preferable.

Fig. 2 shows the course of the bottom quantity and quality for the procedure indicated in Fig. 1. It concerns a ternary distillation of the components A, B, and C in order of

---

*Corresponding author. Tel: +31-53-4893043; fax: +31-53-4893849; e-mail: b.h.l.betlem@ct.utwente.nl

1385-8947/98/$ – see front matter
PII: S1385-8947(98)00118-1

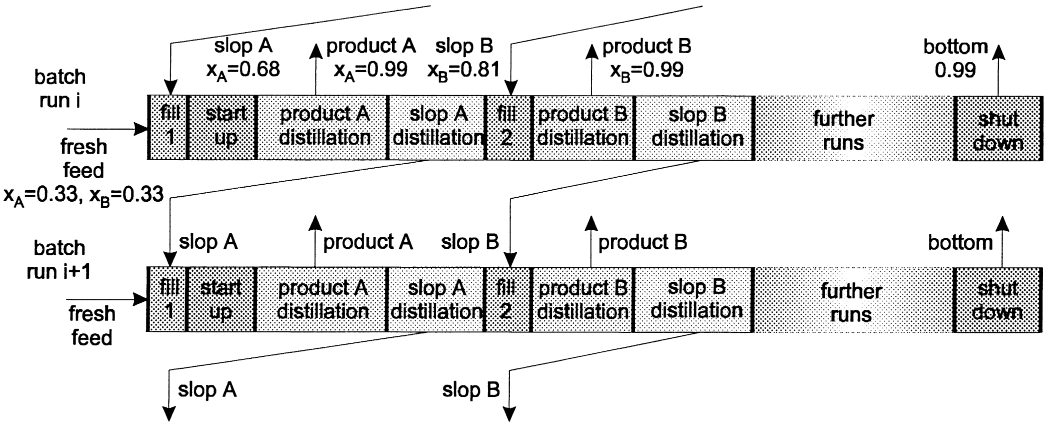


Fig. 1. Phases of batch distillation run. The purities indicated are borrowed from ternary distillation.

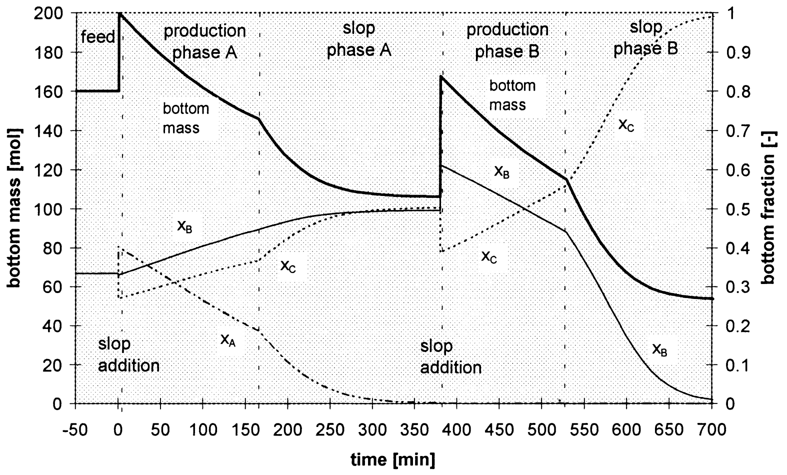


Fig. 2. Typical bottom quantity and quality profile of ternary distillation with slop recycling under constant overhead quality control. Feed: $x_A = x_B = x_C = 0.33$, products: $x_A = x_B = x_C = 0.99$, relative volatility: $\alpha_{AB} = \alpha_{BC} = 1.8$.

decreasing volatility. At starting time the slop of the previous batch is added, resulting in a rise of concentration of A. Next, A is removed, first in product and then in slop. In the mean time the concentrations of B and C increase. After adding the second slop, the concentration of B is augmented. Subsequently, B is produced and finally the bottom is exhausted in B until only product C remains.

For a multi-component separation the following 'profit function' has been chosen.

$$\text{capacity} = \frac{\Sigma\,\text{product amount}}{\Sigma\left(T_{\text{product cut}} + T_{\text{slop cut}}\right)} \tag{1}$$

with the constraints:

- the feed has to be processed completely
- all products, including the residue, should be at purity specification.

This profit function together with the constraints holds for the following assumptions:

- the non-production times and the distillation times are independent
- the market for the main products is a sellers market. Under these conditions production increase leads to more profit than cost reduction
- the unit is a production bottleneck. The combination with the previous assumption requirement implies capacity maximisation
- the profit calculation is independent of product prices and feed costs. By requiring that all feed is processed into products with hard demanded qualities, the mass and component balances leave no further degrees of freedom

The advantage of this profit function is that it is independent of market prices and energy costs. Actually, a part of the optimisation problem is shifted to higher control levels such as plant scheduling and planning. At those levels information about the product purities best suited to the market situation, the product and cost prices, and the realisable unit capacities are available. With this information the assignments of product qualities and quantities (or mean production rate) to the various units can be determined. In a multi-level coordination system the levels should be tuned mutually to achieve the unit goal.

Slop recycling causes mutual dependency of consecutive runs. Therefore, the optimum has been calculated for a cyclic pseudo-steady state. The degrees of freedom are limited to the control strategy for a single run, the ratio of the impurities in the products and, if slop recycling is used, the slop quantity and quality. An important aim of this study is to develop simple models for the scheduling control level. With these models it should be possible to determine the degrees of freedom of slop quantity and quality, and to predict the total production time for optimal operation. First, the best control policy for a single distillation cut will be determined. The policy will depend on the difficulty of the separation. Next, the models for batch distillation with slop recycle under optimal control are build.

2. Literature

In a review paper, Rippin [2] reports about 15 batch distillation optimisation studies done in the period from 1963 to 1980. In all articles the objective was to produce a certain amount of product at requested composition either in minimal production time or with maximal economic profit. Most authors use constant reflux control as a reference policy. Seldom a comparison with constant quality control is made. The question whether optimal policies will show significant improvement over more common methods of operation, therefore, has to be further investigated. The studies are nearly all binary, at constant relative volatility (around 2). The effect of hold-up is usually not taken into account, although it is acknowledged to be important. With the exception of Robinson [3] who mentions the final column state, all authors have neither a bottom product demand nor give information about the state in which the column is left behind. Time advantages of dynamic optimal control up to 13% over constant reflux control and up to 6% over constant quality control are reported. Kerkhof and Vissers [4] report the highest profits. The optimisation objective was maximal profit per unit of time, taking into account the product to feed price ratio and the recharging time. Frequently, the profit improvement was as much as 20% and sometimes much higher. Only Robinson [5] discusses an industrial implementation. The author used a sub-optimal control scheme consisting of constant reflux control which is switched over during the batch to constant top purity control; savings of 13.5% are reached.

Robinson [3] proposed the specific group $\alpha^{n+1}(1-\bar{x}_D)/\bar{x}_D$ to order the results of former studies. The group was derived from the Fenske equation. In his clarifying article, the author concludes that for all low-hold-up operations nearly always a constant quality policy should be used. Strangely enough, in later work this conclusion is never quoted. Kerkhof and Vissers introduce another measure for separation difficulty indicating the desired control strategy. This measure was also derived from the Fenske equation. It was found that for a rather low degree of difficulty, constant quality control is as profitable as dynamic optimal control. For higher degrees of difficulty, dynamic optimal control is always preferable. However, their conclusions depend very much on economic factors. Only very high feed prices or large switch-over times will result in bottom exhaustion.

From 1980 only a few articles were published about batch distillation control without slop recycling. Mostly, the authors introduce a new numerical concept, but the optimisation results do not differ from earlier studies. Hansen and Jørgensen [6] solved the optimisation problem by orthogonal collocation. Farhat et al. [7] claimed that from an industrial point of view, it is desirable to maximise the multiple-fraction separation for a fixed final time. The decision variables are reflux policies for each period and the period switching times. Three predefined shapes of reflux policies are considered: constant, linear increasing, and exponential increasing reflux. The problem is transformed into a non-linear programming problem. Bosley and Edgar [8] used non-linear model predictive control to determine the reflux trajectory. Diwekar et al. [9] based their optimisation on short-cut models. In [10], Diwekar used a combination of the maximum principle and non-linear programming.

The first article concerning slop recycling was already published in 1970 by Mayur et al. [11,12]. The authors describe a single pseudo-steady state example. After 1987 an increasing interest is perceptible. Table 1 shows an overview of the articles published on this subject.

Christensen and Jørgensen [13] studied circumstances under which slop recycling is worthwhile. The authors investigated 31 different cases of binary batch distillation in tray columns and introduced a new degree of separation difficulty q:

$$q=\frac{1}{x_{\mathrm{F}}-x_{\mathrm{B,set}}}\int_{x_{\mathrm{B,set}}}^{x_{\mathrm{F}}}\frac{n_{\min}+1}{n_{\mathrm{ideal}}+1}\,\mathrm{d}x_{\mathrm{B}}\quad\text{with}$$

$$n_{\min}+1=\frac{\ln\left(\left[x_{\mathrm{D,set}}/(1-x_{\mathrm{D,set}})\right]\left[(1-x_{\mathrm{B}})/x_{\mathrm{B}}\right]\right)}{\ln\alpha}\qquad(2)$$

$n_{\min}$ is the minimum number of trays (according to Fenske) required to realise the bottom purity at total reflux, whereas n_{ideal} is the number of ideal trays present in the actual column. q integrates from the feed composition x_{F} to the end bottom composition $x_{\mathrm{B,set}}$, at which exhaustion is reached. The degree of separation difficulty is based on distillation without slop recycle. The authors indicate that when $q > 75\%$ slop recycling offers time savings up to 30%.

Luyben [14] determined the optimum number of stages and the optimum reflux ratio. For the optimum criteria the capacity factor is used. This factor is defined as the total specification products produced, divided by the total time of the batch. Two different control strategies are tested:

Table 1
Literature overview concerning batch distillation with slop recycling

Authors	Problem specifications				Tray hold-up	Mixture	Reflux strategy/mathematical method	Time advantage
	$X_{D,A}$	$X_{D,B}$	n	α				
[11]	0.85	0.90	5	2	No	Binary	Do Pontryagin+CR	3% over no slop; DO over CR: 4%
[13]	0.75–0.95	0.50–0.94	2–9	1.65–2.25	No	Binary	DO / orthogonal collocation	0–25% over no slop in one case: 92%
[14]	0.95	0.95	20+40+60	9/3/1	Yes	Ternary	CR+kind of CQ	CR maximal 15% better
[1]	0.95+0.99	0.95+0.99	40	4/2/1+9/3/1	Yes	Ternary	6 CR strategies	Up to 35% over total slop recycle
[15]	0.85–0.95	0.99	4+5	C4–C5–C6	2%	Ternary	CR (main: 3 × CR, slop: 1 × CR) / SQP	More than 45% over no slop
[16]	0.95	0.95	6	3	Yes	Binary multi.	CR, 2 level optimisation. / SQP	Goal: optimisation framework
[17]	0.80–0.97	0.80–0.85	5–30	2×Antoi ne	No	Ternary	CR optimisation of slop-cut addition / NLP	1.5% and 5.5% over separate distillation of slops

$x_{D,A}$, $x_{D,B}$ – product specifications, n – number of trays, α – relative volatility.
DO – dynamic optimisation, CR – constant reflux, NLP – non-linear programming, SQP – successive quadratic programming.

constant reflux and variable reflux. The variable reflux controller increases the reflux flow when the distillate purity is above a specified value. It acts as a kind of quality controller. For the 40-tray column the differences are small. But for the 20-tray column when the difficulty of separation is high, then the constant reflux ratio strategy yields a 15% higher capacity factor. The variable distillate operation reduces the batch time, but decreases the fresh feed since larger slop cuts are produced.

Mujtaba and Macchietto [15] developed an optimal control strategy which allows calculating simultaneously the amounts and compositions of the recycle cuts and the reflux ratio profiles to yield a minimum batch cycle time. The product cut has been divided into three piecewise constant reflux intervals. For this cut the three reflux ratios and three switching times are determined. They show that an adapted degree of separation difficulty q can be used in deciding whether a recycle is sensible. When $q = 56\%$ the proposed optimal recycling strategy gives an overall batch time saving of more than 45%.

Mujtaba and Macchietto [16] developed a structure permitting the calculation of optimal policies for each alternative operation with consideration of multi-period objectives and constraints. A two-level optimal control formulation is proposed. The multi-period operation is decomposed into a sequence of independent optimal control problems. The outer level maximises the profit function, whereas the inner loop solves the minimum time problem. The binary runs are decoupled by assigning prices to the slop cuts (SC). The SC produced gains money, whereas the SC used, which is a part of B_{initial}, costs money. The constrained non-linear optimisation problem is solved using a successive quadratic programming technique. Their approach is very powerful. A disadvantage is that prices for all intermediates are required.

Bonny et al. [17] do not use the pseudo-steady state approach. They present a general model which allows the calculation of the proportion of each slop cut to be added to each initial load and to each new batch. The authors illustrate their approach by distilling a fixed quantity with a limited reboiler capacity. This requires multiple runs. Future fresh feed is not taken into account. Partial slop addition to the next runs is shown to be more profitable than other strategies. They also conclude that a fixed reflux ratio appears to be nearly as good as other variable reflux policies (2.3% difference). However, mixing slop cut with fresh feed to reduce the degree of separation difficulty is the basic principle of slop recycle. This principle does not agree with fixed time or fixed quantity problems.

The literature clearly indicates that slop recycling can be profitable. In nearly all articles constant reflux control is applied. Then the reflux settings during the different product and slop cuts are the degrees of freedom. Different recycle strategies are applied. However, the articles give no insight, why a particular control policy is applicable in a particular context. Nearly all of them pose specific examples and only one article [13] discusses a broad range of circumstances.

3. Optimal control policy for a single cut

For a single distillation cut the relative performance of dynamic optimal control has been compared with control policies which can be implemented much easier in practice. It is important to clarify which simple policy performs best under certain circumstances, especially, which performs best in case of slop recycling. In the literature, the so-called difficulty of separation is used to characterise a batch distillation. However, this measure is only based on the start conditions of a distillation cut. Therefore, a second independent measure is introduced which is based on the column state at the end of a cut: the degree of exhaustion. The combination of the two measures gives a better understanding about why and when a method is applicable. The following basic control policies are investigated:

- constant reflux control: CR
- constant quality control: CQ
- dynamic optimisation according to the principle of Pontryagin: DO.

To control a single batch distillation run two manipulated variables are available: the reflux ratio and the boil-up energy. The energy input is used to set the column load and the reflux ratio is used to control the product quality. Optimal reboiler heat input is usually taken to be the heat supply generating a vapour load close to its maximum value without sacrificing too much tray efficiency. Depending on the tray type this transition is sharp or more gradual. As in most optimal control studies (except in Bosley and Edgar [8]), here the boiling-up rate was taken constant. The tray-to-tray model is added in Appendix A. The equations necessary to solve the dynamic optimisation according to the Pontryagin's maximum principle, together with the solution procedure can be found in Betlem [18].

Fig. 3 shows the maximum feasible top product as a function of the relative volatility for the three control policies. The maximum production for CR is equal to those for DO, and always higher than those for CQ. The figure indicates four production zones. In zone 1 (up to $\alpha = 1.6$ for this column) no top product at the required purity can be distilled. The introduction of slop recycle will not change this. But, in zone 2 production of the required top product is possible. However, the bottom cannot be sufficiently exhausted to obtain the required bottom composition. Slop recycling is necessary for DO, CR, as well as CQ to realise this. Also in zone 3 for CQ slop recycling is still required. In zone 4 with a single cut without slop recycling the top as well as bottom product can be distilled on specification.

Thus, batch distillation runs can be characterised by the difficulty or capability of an installation to process a certain

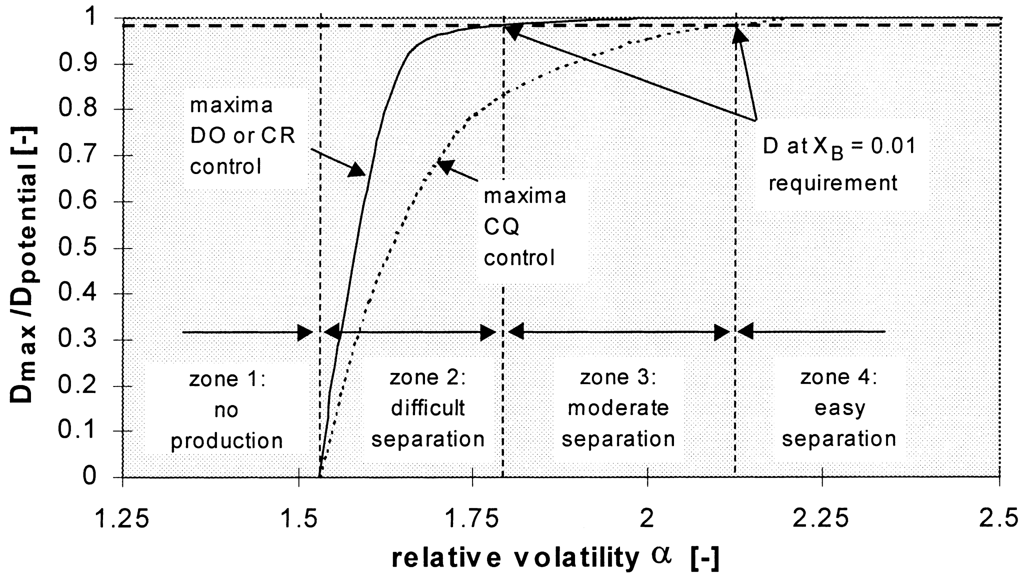


Fig. 3. Production zones for column (21 trays ε_v=0.53) with top and bottom specification ($x_{D,set}$=0.99 and $x_{B,set}$=0.01) under dynamic optimal (DO), constant reflux (CR), and constant quality (CQ).

product quality from a feed and by the extent the feed is exhausted. Therefore, two measures are introduced to describe these characteristics. This is the basis on which the control strategies are distinguished. To indicate the desired control strategy Kerkhof and Vissers [5] used the difficulty of separation. The definition is

$$\sigma_{\text{dif}} = D_{\text{potential}} - \frac{D_{\text{maxCQ}}}{D_{\text{potential}}} \times 100\% \tag{3}$$

$D_{\text{max CQ}}$ is the maximal production under constant quality control and $D_{\text{potential}}$ is the theoretical maximal production. The formula expresses the maximal capability of CQ control, which is reached if the reflux ratio goes to infinity, still producing the demanded top purity. This relation can be described by means of the Fenske equation. Substitution of the theoretical maximal amount of component x, and the maximal amount of distillate under CQ, the definition results in

$$\sigma_{\text{dif}} = \frac{x_{\text{D,set}} - x_{\text{B,0}}}{x_{\text{B,0}}(1 - x_{\text{D,set}})} \times \frac{1}{\alpha^{\varepsilon_{\text{column}}(n+1)} - 1} \times 100\% \tag{4}$$

The degree of separation difficulty is only dependent on the initial bottom fraction, the desired top purity and the column capability. The desired final column state is not included. Therefore, a supplementary degree of difficulty is necessary for describing the exhaustion at the end of the production. The degree of exhaustion definition becomes

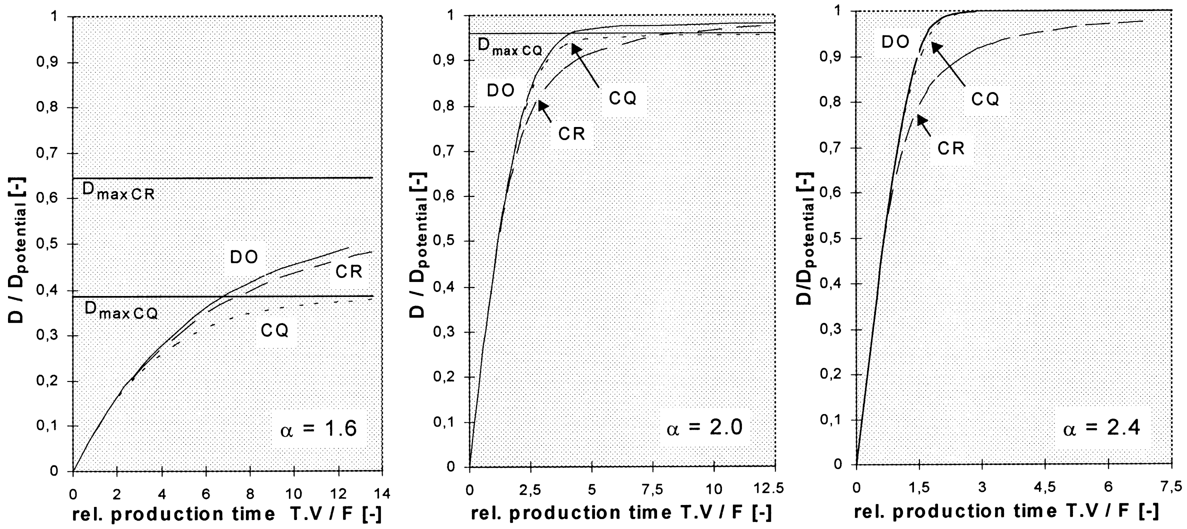
$$\sigma_{\text{exh}} = \frac{D}{D_{\text{maxCR}}} \times 100\% \tag{5}$$

which describes exhaustion as the ratio between the production realised D and the maximal exhaustion under CR: $D_{\text{max CR}}$ (which is equal to maximal exhaustion under DO).

For three areas of separation difficulty Fig. 4 shows the maximal product amount which can be realised for a certain production time under CQ, CR or DO. Note that a curve does not show the production course of a run. Production depends on relative volatility which characterises the difficulty of separation. For every relative volatility the maximal feasible production is obtained when the production time goes to infinity. These maximal production values correspond to the values along the curves in Fig. 3. However, when the production time goes to infinity, the production rate goes to zero.

- Difficult separation ($\alpha = 1.6$ in Fig. 4: $\sigma_{\text{dif}} = 40\%$). At high degrees of difficulty for all control strategies the production is limited. DO and CR achieve nearly the same capacity. Both use a high reflux rate. Note that at an exhaustion of $D/D_{\text{potential}} = 30\%$ ($\sigma_{\text{exh}} = 50\%$) DO uses 15% less time than CR to produce the same product amount. For $D/D_{\text{potential}} < 25\%$ ($\sigma_{\text{exh}} < 40\%$) CQ has equal production. At long production times the product amount lags behind with up to 40%.
- Moderate separation ($\alpha = 2.0$ in Fig. 4: $\sigma_{\text{dif}} = 3\%$). At medium degrees of difficulty CQ has a lower production limit than CR and DO. But, even at relatively high exhaustion (above 90%) the performance of CQ is equal to DO and much better than CR.
- Easy separation ($\alpha = 2.4$ in Fig. 4: $\sigma_{\text{dif}} = 0.4\%$). At low degrees of difficulty CQ becomes equal to DO. Only for $\sigma_{\text{exh}} < 60\%$ CR has equal production.

DO always has the highest production for every production time. However, if σ_{dif} or σ_{exh} are restricted then the improvement of DO with respect to CR or CQ is small (see Fig. 5). CR can be used for nearly every situation with a



a. difficult separation *b. moderate separation* *c. easy separation*

Fig. 4. Relative maximal production vs. the relative batch time for DO, CR and CQ control if α=1.6, 2.0, and 2.4 ($x_{D,set}$=0.99, x_F=$x_{B,0}$=0.5, n=21 and ε_v=0.53).

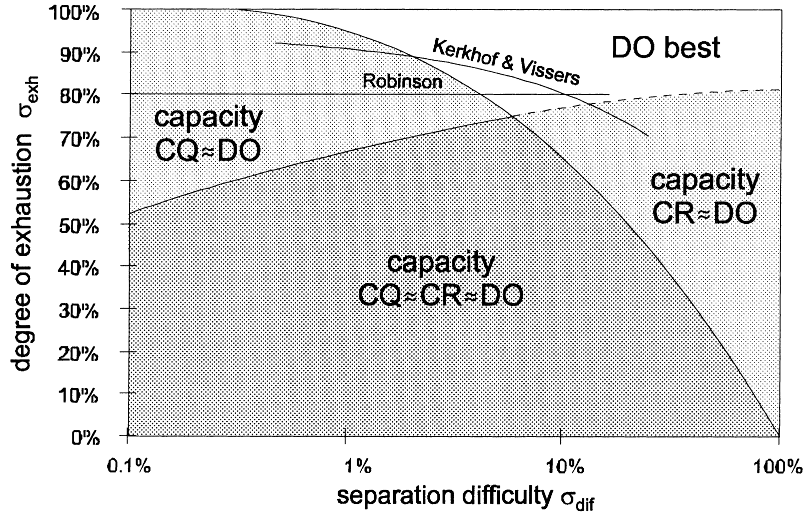


Fig. 5. Area for which CQ and CR perform 2% less in comparison with DO.The investigated areas by Robinson and by Kerkhof and Vissers are indicated.

limited bottom exhaustion: if σ_{exh} remains below 60–70%. CQ can be used if either the difficulty of separation or the degree of exhaustion is low.

Kerkhof and Vissers [5] recommended the following control policies: if $\sigma_{dif} \leq 4\%$ then CQ can be applied, if $4\% < \sigma_{dif} < 15\%$ then DO exceeds CQ or CR, and if $\sigma_{dif} \geq 15\%$ then CR is applicable. Their simulations are indicated by a curve in Fig. 5. The authors did not consider the exhaustion of the bottom since an economic criterion was applied. Consequently, a more difficult separation has an optimum at less exhaustion. The recommendations correspond reasonably well to the performance areas in the diagram. The study of Robinson [3] quoted in the literature section is indicated in Fig. 5 as well.

4. Optimal operation of batch distillation with slop recycling

The optimal operation for batch distillation with slop recycling is determined by varying the degrees of freedom in the case of a cyclic pseudo-steady state operation. Binary batch distillation with slop recycling has two degrees of freedom, which are the independent variables in the optimisations. In the case of CQ the degrees of freedom are the slop quantity SC and slop quality x_{SC}. In case of CR it is more convenient to use the reflux ratios during the product and the slop cut. The resulting optimal exhaustion after the product cut and the production time of the product cut as well as of the slop cut are dependent variables. Thus, every

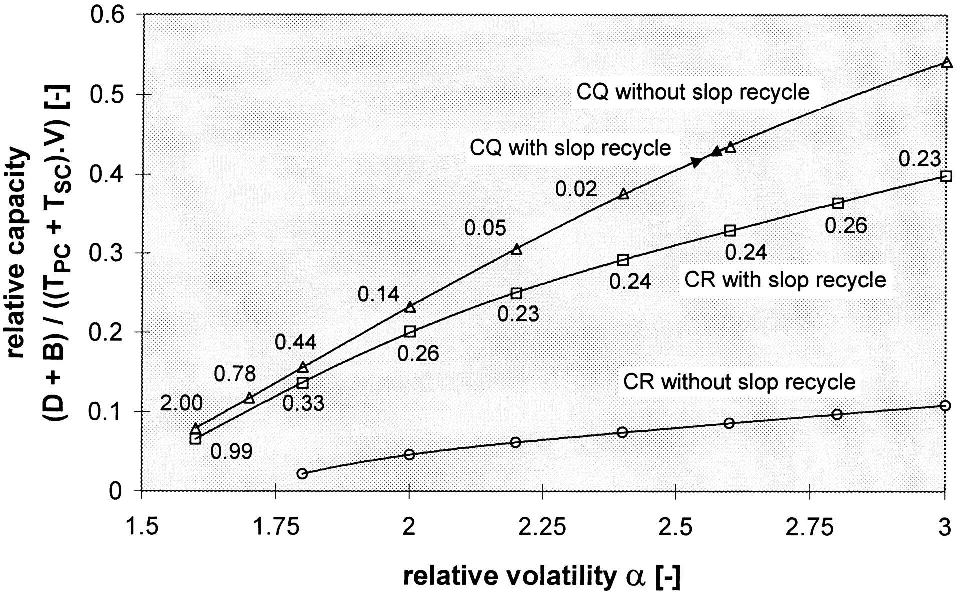


Fig. 6. Comparison between CR, CR + slop recycling and CQ control. The numbers along the curve indicate the relative slop amount (SC/F). Conditions: $1.6 \leq \alpha \leq 4$, $x_F = 0.5$, $x_{D,set} = 0.99$, $x_{B,set} = 0.01$, $\varepsilon_V = 0.53$, $n = 21$.

optimal point is the result of an extensive calculation. Two control policies are compared: CR and CQ. Afterwards the results are checked with the policy criteria developed in the previous section. The results are expressed in the obtained relative capacity.

$$\text{Relative capacity} = \frac{\text{product amount}}{\text{production time}} \times \frac{1}{V} = \frac{D+B}{T_{PC}+T_{SC}} \times \frac{1}{V} \quad (6)$$

The binary batch distillation delivers two products: the distillate product D and the bottom product B. If the feed and the product requirements are fixed, the pseudo-steady state operation fixes the product amounts D and B. The total batch time required is the sum of the product distillation time and the slop distillation time. As the vapour flow is linear with the production rate, the vapour flow V is used to normalise the profit. Thus, optimising the capacity means minimising the total batch time, being the only dependent variable in the profit function. In the simulation studies the relative volatility α is the most important parameter to vary the degree of separation difficulty over a wide range. For top and bottom product high quality requirements apply: $x_{D,set} = 0.99$ and $x_{B,set} = 0.01$. Usually, the feed purity x_F is 0.50. But in a few cases other feed compositions are used to investigate the feed composition dependency of a result.

In Fig. 6a comparison between the CR and CQ policy is made. Every capacity result (point in graphic) is the maximal production capacity under pseudo-steady state circumstances regarding the two product purity demands. From the simulations it appeared that for high αs, the optimum is sensitive to the slop amount and insensitive to quality fluctuations, whereas for relatively low αs the opposite applies.

For difficult and moderate separations the capacity for CQ is about 20% higher than for CR with slop recycling. For easy separations the difference in control policy can exceed 35%. If CQ is applied, only below a particular difficulty of separation slop recycle is advisable (here at $\alpha < 2.55$). Coming from $\alpha = 1.6$ for higher αs less and less slop is required until the amount of slop becomes zero. Above $\alpha = 2.25$, (according to Fig. 3) slop recycling is not required to fulfil the bottom composition, but still does improve the production rate. If CR is applied the situation is different. Even for relatively high αs, slop recycle substantially improves the capacity of CR. Bottom exhaustion in one run takes much time for CR. Slop recycling is required to relieve the bottom exhaustion.

Fig. 7 shows the production progress for a pseudo-steady state run with slop recycle. The relative volatility has been chosen almost at its minimum value allowing 99% top purity: $\alpha = 1.6$. This gives the highest degree of separation difficulty. The capacity under CQ is higher, despite large exhaustion during the slop cut. At the end of the slop phase the relative production increase is almost zero (see Fig. 7). The improvement in time of about 17% is evenly partitioned over the product and the slop cut. The results agree with the single-run conclusions. For the product cut under CQ the degree of exhaustion $\sigma_{exh} = 20\%$ and the degree of separation difficulty $\sigma_{dif} = 15\%$. Then CR performs as well as CQ. However, during the slop cut $\sigma_{exh} = 99\%$ and $\sigma_{dif} = 0.4\%$. Under these circumstances, CQ is superior to CR control. On the conditions applying during the product cut and the slop cut, DO can gain a potential capacity which lies only a fraction higher than by using CQ.

Fig. 8 shows the simulation results under CQ with slop recycle for a broad range of the degree of difficulty (σ_{dif}).

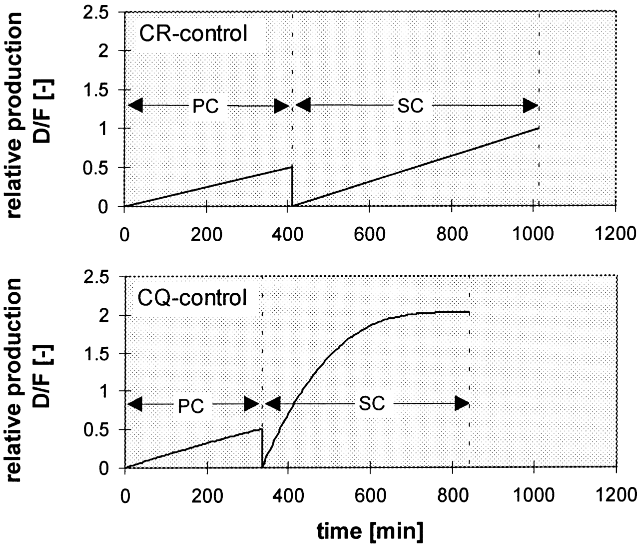


Fig. 7. Distillation production course under CR, respectively, CQ control for difficult distillation; PC = product cut and SC = slop cut. Conditions: $\alpha = 1.6$, $x_F = 0.5$, $x_{D,set} = 0.99$, $x_{B,set} = 0.01$, $\varepsilon_V = 0.53$, $n = 21$.

The range of relative volatility goes from 1.6 (very difficult) until 4.0 (very easy separation). Also, the feed composition and the final bottom composition have been varied. The degree of exhaustion (σ_{exh}) of an optimal operating point in the figure is fixed by optimisation and, therefore, is a dependent variable. The curve in the figure bounds the optimal control policy areas as derived in the previous chapter. Nearly all optimal distillation cuts follow a fixed patron. They are situated in the area where constant quality control is almost as good as dynamic optimal control. Surprisingly, all product cut points are arranged along this boundary curve. At high degrees of difficulty exhaustion is avoided. The difficulty of separation is reduced by the applied slop quantity and quality. Apparently, maximal capacity is achieved when the product is gained and no sacrifices are made towards an optimal policy. The slop cut points are grouped around the maximal exhaustion as the bottom requirement of 1% in the light component should be met. Now high degrees of difficulty are avoided. Three slop cut points have a significantly lower σ_{exh}. These cases concern easy separations ($\alpha = 2.4$). Then, after product distillation, the bottom is already nearly exhausted. Slop recycling combined with constant quality control offers a control policy comparable with optimal control. This important conclusion does not correspond with the control strategy suggested in the literature.

5. Scheduling models

It is important to have scheduling models which enable to determine the optimal degrees of freedom and total production time without elaborate and extensive computer simulations. The models for the optimal values are based on specific measures which make them rather universal. It appeared that the product-to-slop amount ratio for the actual feed quantity and quality can be calculated by means of the slop indication measure σ_{slop}. This measure corresponds to the specific measure q defined by Christensen and Jørgensen [13]. In their study they used a tray efficiency of 100%. The column efficiency ε_{column} is incorporated into the equation:

$$\sigma_{slop} = \frac{1}{x_F - x_{B,set}} \int_{x_{B,set}}^{x_F} \frac{n_{min} + 1}{\varepsilon_{column} n + 1} dx_B \times 100\% \qquad (7)$$

Note that σ_{slop} is based on the feed composition. This measure can be used when the slop amounts are not known yet. It can deal with non-steady state situations, in which the feed alters or when sufficient slop is not available.

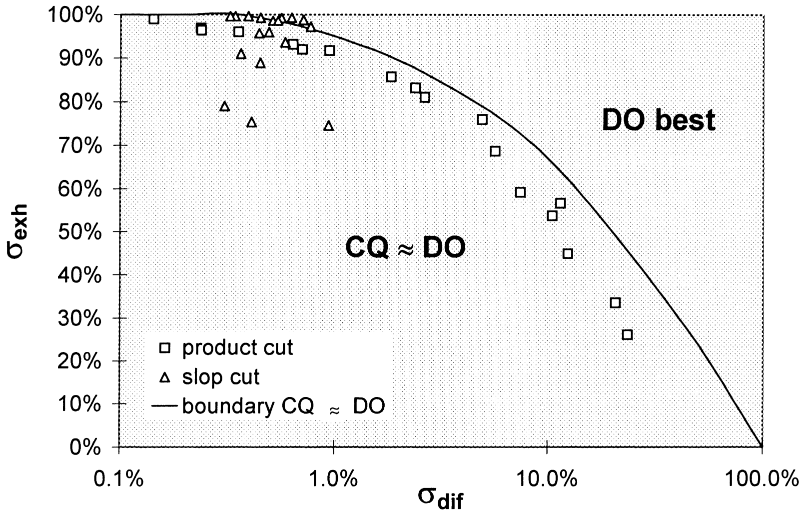


Fig. 8. Position of σ_{dif} and σ_{exh} combinations of product and slop cuts of 30 simulations with slop recycling. Conditions: $1.6 \leq \alpha \leq 4$, $x_F = 0.5, 0.6, 0.8$, $x_{D,set} = 0.99$, $x_{B,set} = 0.01, 0.05$, $\varepsilon_V = 0.53$, $n = 21$.

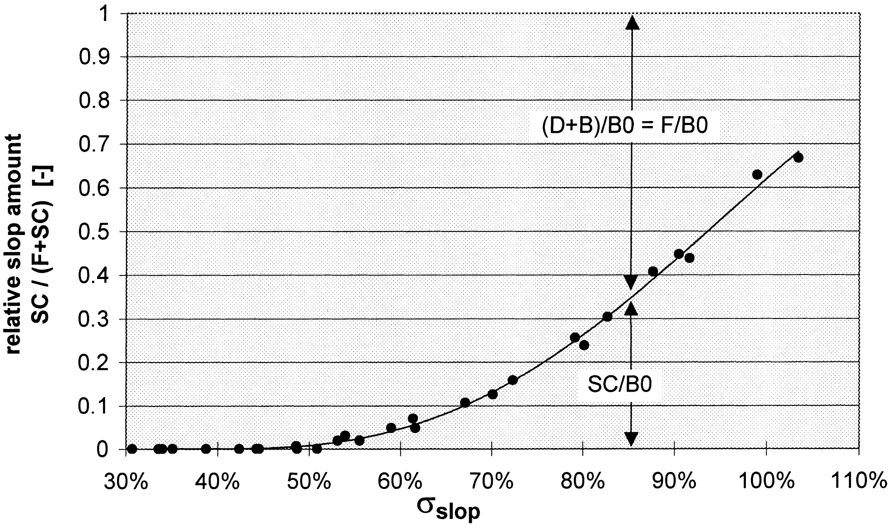


Fig. 9. Relation between the slop indication measure σ_{slop} and the optimal feed to slop ratio. The results of 32 optimisations are depicted. Conditions: $1.6 \le \alpha \le 4$, $x_F = 0.5, 0.6, 0.8$, $x_{D,\text{set}} = 0.99, 0.98$, $x_{B,\text{set}} = 0.01, 0.05$, $\varepsilon_V = 0.53$, $n = 21$.

Fig. 9 shows the relation resulting from computer simulations. Surprisingly, the curve is independent of relative volatility, feed composition and product and bottom requirements. For all simulations the degrees of freedom are determined in such a way that the degree of difficulty and the degree of exhaustion are tuned according to a fixed patron as described in Fig. 8. The transitional area where slop recycle becomes useful is located around 60%. This percentage corresponds to the results of Christensen and Jørgensen [13], although they used CR instead of CQ and in that study column efficiency was not taken into account.

Kerkhof and Vissers [5] introduced a measure to indicate the difficulty of separation. It appeared that the slop amount as well as the slop total time necessary for product and slop distillation can be estimated by means of a linear relation of the average separation difficulty: $\sigma_{\text{dif,avg}}$, which is the average of the specific measure defined by Kerkhof and Vissers taken over the product cut.

$$\begin{aligned}\sigma_{\text{dif,avg}} &= \frac{1}{x_{B,1} - x_{B,0}} \int_{x_{B,0}}^{x_{B,1}} \sigma_{\text{dif}} \mathrm{d}x_B \times 100\% \\ &= \frac{x_{D,\text{set}}}{1 - x_{D,\text{set}}} \left(\frac{1}{x_{B,1} - x_{B,0}} \times \ln\left(\frac{x_{B,1}}{x_{B,0}}\right) - \frac{1}{x_{D,\text{set}}} \right) \\ &\quad \times \frac{1}{\alpha^{\varepsilon_{\text{column}}(n+1)} - 1} \times 100\% \end{aligned} \quad (8)$$

In this definition $x_{B,0}$ is the initial bottom fraction and $x_{B,1}$ is the bottom fraction after product distillation. Note that $\sigma_{\text{dif,avg}}$ is based on the initial bottom composition after addition of the slop and on the composition at the end of the product cut. Therefore, to use this measure the degrees of freedom should be known.

Fig. 10 shows the relations determined from 30 simulation optimisation results. The relation is independent of feed composition and relative volatility, but depends on the product quality requirement. It is convenient that production time can be described by a linear relation in a specific measure. The straight line can be characterised by two parameters: a slope and an abscissa. These two parameters can be adapted from measurements for a particular column and different products. If on-line adaptation is made according to the reported progress (with forgetting factor), the scheduler can be updated to the current column performance after every run.

The three derived schedule models can be applied according the following procedure:

- Partition the batch distillation in a sequence of pseudo-binary separations and determine from measurements the column separation capability $\alpha^{\varepsilon_{\text{column}}(n+1)}$ for these separations.
- Check for each binary separation whether the combination of degree of difficulty and exhaustion lies within the area of constant quality control (Fig. 8). If not, then slop recycling is required. The position in the figure indicates whether the difficulty or exhaustion is limiting.
- Determine the optimal slop amount by means of σ_{slop} (Fig. 9). The slop amount can be corrected for trends in the feed quantity and quality. This is worked out in Betlem [18].
- Theoretically, the optimal slop composition can be calculated by combining the relations from Figs. 9 and 10(A). These figures offer two independent relations for two unknown degrees of freedom. Practically, it is more convenient to choose a fixed composition positioned between the feed composition and product requirement. The sensitivity of the optimum for slop composition is rather small.
- Next, the production time prediction that depends on the settings of the degrees of freedom can be determined. The time can be directly derived by combining Figs. 9

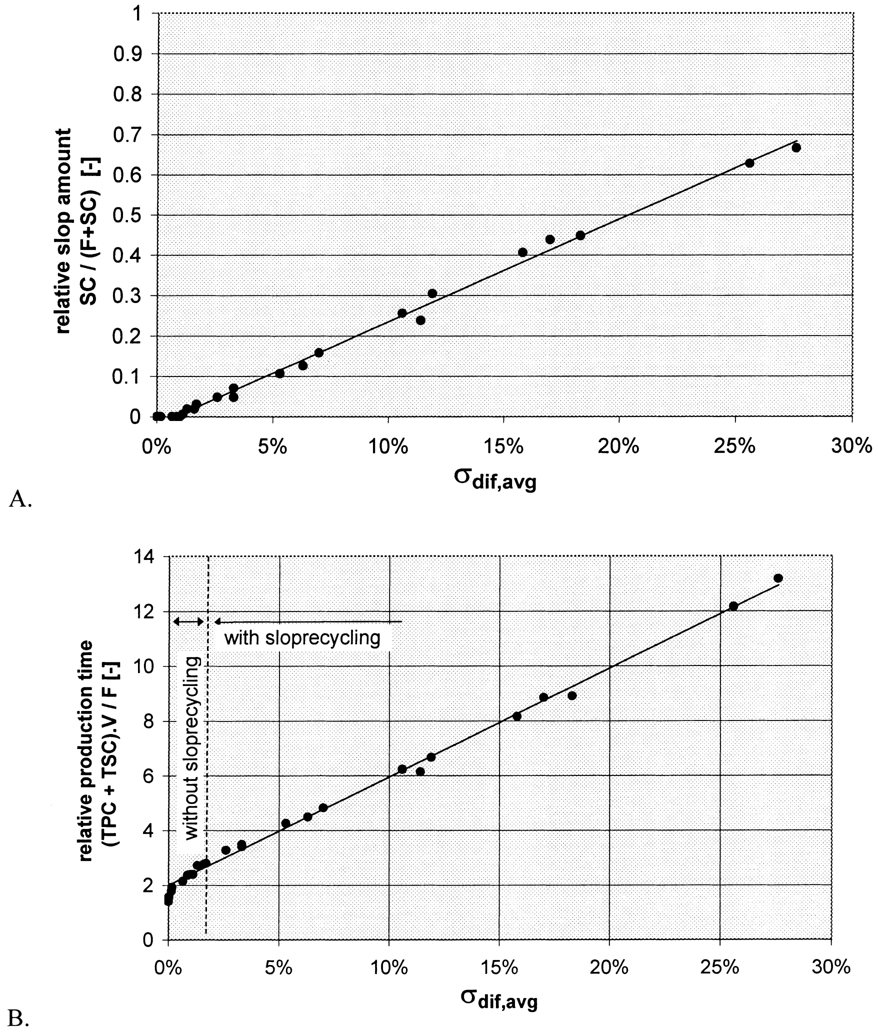


Fig. 10. The relations between the average separation difficulty $\sigma_{\text{dif,avg}}$ and the relative slop amount as well as the total production time derived from 30 optimisation simulations. Conditions: $1.6 \leq \alpha \leq 4$, $x_F = 0.5, 0.6, 0.8$, $x_{D,\text{set}} = 0.99$, $x_{B,\text{set}} = 0.01$, $\varepsilon_V = 0.53$, $n = 21$.

and 10(A) and (B). When a fixed slop quality is used, then the average separation difficulty should be calculated from mass balances and Fig. 10(A) is not necessary.

6. Ternary distillation

Batch distillation is nearly always a multiple component separation. In this study the separations of three components A, B, and C is considered. A is the most volatile and C the least volatile component. Two separations are necessary: A from B, and B from C. Both separations are composed in a product and a slop distillation. The state in which a column is left behind after a separation forms the initial state of the next run. The runs are coupled in a forward manner. The only linking factor is the impurity ratio (x_A/x_C) of product B, which offers an extra degree of freedom for optimisation. When this ratio is assigned, consequently, the amount of the most volatile component in the feed (bottom residue after slop distillation of A) is fixed. During the next run all relatively volatile components in the bottom residue will come over the top. When the bottom contains many of these components, then the next less volatile product probably cannot be produced on specification. Also, if only a small amount is left, then this restricts the possibilities to produce at high purity. When the quality demand is 0.99, then the margin of 0.01 must be divided between the more volatile impurities (here: A) and the less volatile impurities (here: C).

The influence of a ternary distillation on distillation capacity, the settings of the degrees of freedom, and the scheduling models are examined. In the product cut of component A, component C goes along with the bottom whereas in the second product cut concerning component B, the relatively volatile component A comes over the top. The composition which remains in the column after a run determines the separation difficulty of the sequential run. The α combinations used are: $\alpha_{AB} = 1.8/\alpha_{BC} = 1.8$, $\alpha_{AB} = 2.2/\alpha_{BC} = 1.8$, $\alpha_{AB} = 1.8/\alpha_{BC} = 2.2$, $\alpha_{AB} = 2.2/\alpha_{BC} = 2.2$. These combinations correspond to

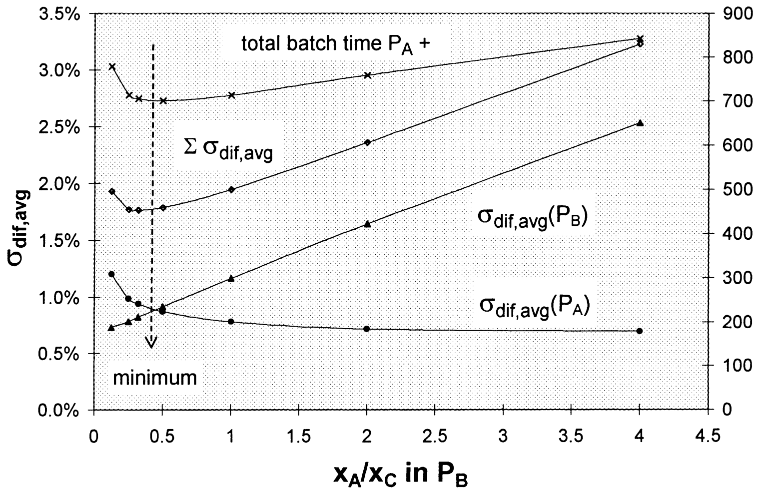


Fig. 11. Relation between the position of the minimal batch time and the degrees of difficulty of the single runs. The degrees of difficulty are a function of the impurity ratio x_A/x_C. Conditions: $\alpha_{AB}=\alpha_{BC}=1.8$; $x_{F,A}=x_{F,B}=0.33$; $x_{D,A}=x_{D,B}=x_{D,C}=0.99$, $\varepsilon_V=0.53$, $n=21$.

four different sequences in degrees of separation difficulty: difficult/difficult, easy/difficult, difficult/easy, and easy/easy, respectively.

For optimal multi-component separation the same main rule as for binary separation applies: spreading of difficulty of separation over a distillation run. The optimal slop quantity and quality together with the optimal impurity ratio are determined in such a way that constant quality control for each cut approaches dynamic optimal control. The difficulties of separation are balanced between production and slop cuts and between the different products.

When the first separation is relatively difficult (α_{AB} is low) then the optimal impurity ratio is nearly equal to the volatility ratio. However, when the second separation is relatively difficult, then A should be relatively under-represented in B. The reason for this phenomenon is explained in Fig. 11. If the impurity ratio x_A/x_C in product B increases, on the one hand the difficulty of distillation of product A drops, as more A is allowed in the bottom, whereas on the other hand the difficulty of distillation of product B rises due to the large proportion of A in distillate B. The relatively small fraction of A in the bottom can be more easily compensated for by a larger slop recycle than a relatively large fraction of A in the top. Instead of $x_A/x_C=\alpha_{AB}/\alpha_{BC}=1$ the optimum is $x_A/x_C=0.5$ (point also indicated in Fig. 11). Fortunately, the sensitivity for the optimum is rather low. In practice the impurity ratio can be taken to be equal to the volatility ratio with, possibly, a correction for high volatility ratios.

For ternary distillation the dependence of the required slop quantity on the slop identification measure remains the same. Fig. 12 shows both the ternary and binary results. Also, the relation between average separation difficulty and batch time remains linear and has about the same slope as binary distillation. The independent axis is the mean of the two average separation difficulties. The batch time of a difficult separation followed by an easy separation appeared to be practically equal to the inverse case: the ternary distillation with $\alpha_{AB}=1.8/\alpha_{BC}=2.2$ took 508 min whereas $\alpha_{AB}=2.2/\alpha_{BC}=1.8$ took 517 min. The difficul-

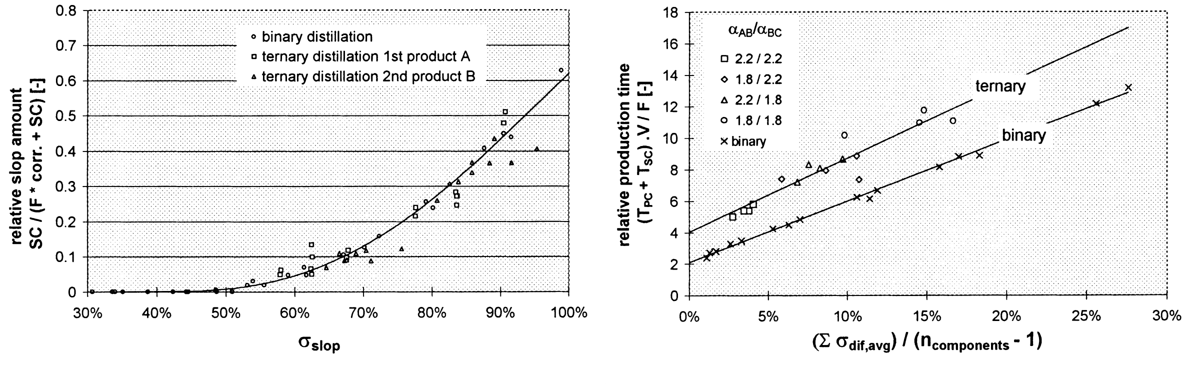


Fig. 12. Relation between the slop indication measure σ_{slop} and the optimal feed-to-slop ratio, and the relation between the average separation difficulty $\sigma_{dif,avg}$ and the total production time ratio. Conditions: 4 α combinations $\alpha_{AB}=1.8/\alpha_{BC}=1.8$, $\alpha_{AB}=2.2/\alpha_{BC}=1.8$, $\alpha_{AB}=1.8/\alpha_{BC}=2.2$, $\alpha_{AB}=2.2/\alpha_{BC}=2.2$; 4 feed fraction combinations: $x_{F,A}/x_{F,B}=0.25/0.25$, $x_{F,A}/x_{F,B}=0.25/0.5$, $x_{F,A}/x_{F,B}=0.33/0.33$, $x_{F,A}/x_{F,B}=0.5/0.25$; product: $x_{D,A}=x_{D,B}=x_{D,C}=0.99$, $\varepsilon_V=0.53$, $n=21$.

ties are always distributed over the distillation runs such that a minimum total time results. The dependent abscissa at $\sigma_{\mathrm{dif,avg}} = 0$ of the ternary distillation is twice the value of the binary distillation because a ternary batch needs two distillation runs instead of one.

7. Influence of hold-up

Tray and condenser hold-ups cause two types of delays. The first type is the top composition lag with a medium range time constant. The second type is the delay obtained during start-up and by the transitions between the product and slop phases. Hold-up reshuffling takes time and increases the degree of separation difficulty. At the end of the batch run the tray and condensor contents are assumed to remain in the column (bubble cap tray), while the bottom is emptied. The top accumulator contents (1% of feed amount) are always added to the slop.

The capacity decrease due to hold-up (Fig. 13) can be considerable. When the tray contents are 25% of the feed amount, for low α the capacity decreases with 25%. In the case of hold-up, the degree of separation difficulty α_{dif} and the degree of exhaustion σ_{exh} are tuned in the same way as separation without hold-up. Then CQ performs almost as good as DO control. A larger hold-up has the same effect on the relative capacity. Therefore, the influence of hold-up on relative capacity decrease is independent of relative volatility.

Specific measures (slop indication measure σ_{slop} and average separation difficulty $\sigma_{\mathrm{dif,avg}}$) are used to calculate the required relative slop amount (SC/B$_0$) and the total relative batch time ($(T_{PC} + T_{SC}) \times V/F$) The relation between the slop indication measure and the required slop amount is shown in Fig. 14. The hold-up increases the degree of difficulty which shifts the curve in Fig. 14 to higher slop amounts. Also in Fig. 14 the relation between the average separation difficulty and the total batch time is shown. A slop increase results in a longer batch run time. If slop recycling is used then the predicted time still corresponds to a nearly linear relation with average separation difficulty $\sigma_{\mathrm{dif,avg}}$. The figure shows that the slope of the relation remains the same.

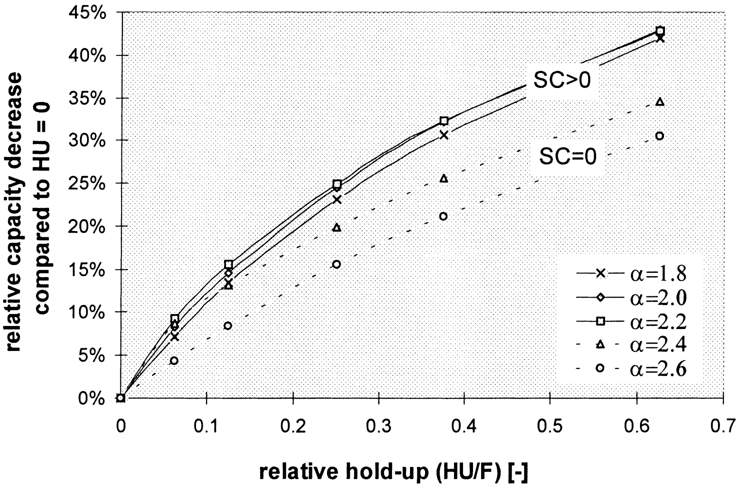


Fig. 13. Relative capacity decrease as a result of hold-up. Conditions: $0 \le \mathrm{HU_{rel}} \le 0.5$, $1.8, \le \alpha \le 2.6$, $x_F = 0.5$, $x_{D,\mathrm{set}} = 0.99$, $x_{B,\mathrm{set}} = 0.01$, $\varepsilon_V = 0.53$, $n = 21$.

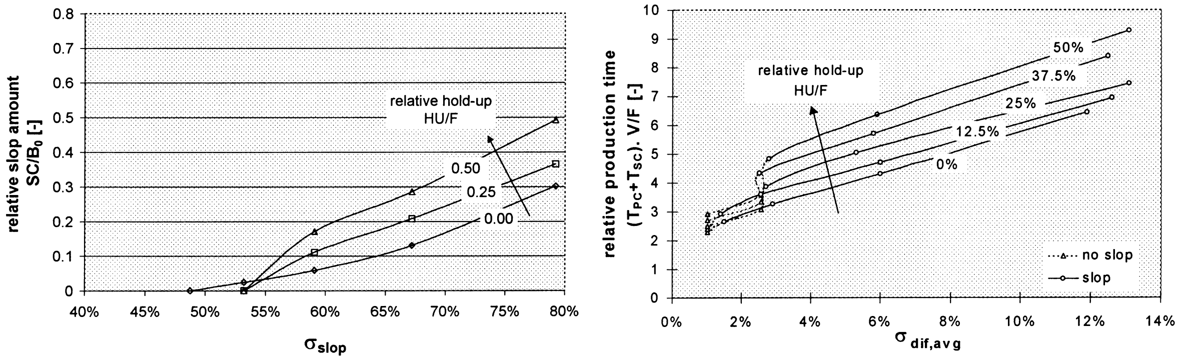


Fig. 14. Influence of the slop indication measure σ_{slop} on the optimal feed to slop ratio, and the influence of the average separation difficulty $\sigma_{\mathrm{dif,avg}}$ on the total production time due to variations in the relative hold-up based on 15 simulations. Conditions: $\mathrm{HU_{rel}} = 0$–50%, $1.8 \le \alpha \le 2.6$, $x_F = 0.5$, $x_{D,\mathrm{set}} = 0.99$, $x_{B,\mathrm{set}} = 0.01$, $\varepsilon_V = 0.53$, $n = 21$.

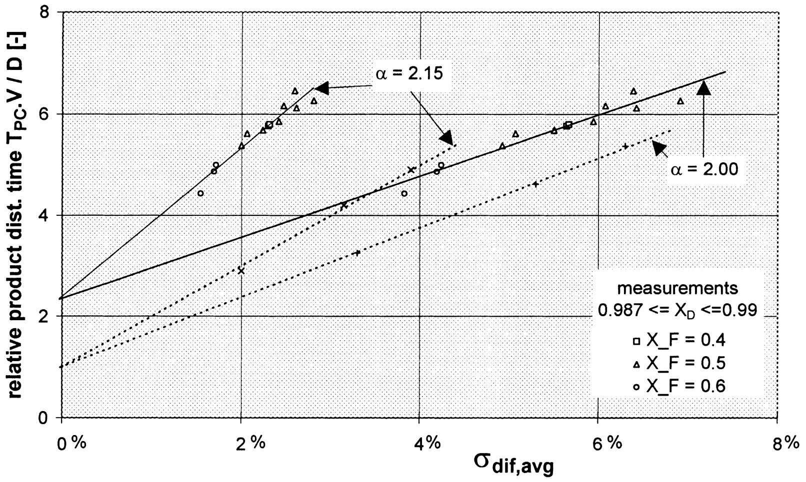


Fig. 15. Measured relation between degree of difficulty $\sigma_{dif,avg}$ and the product cut distillation. The theoretical lines (dotted lines) are also indicated.

8. Experimental results for binary distillation

The average difficulty and production time found from simulations is experimentally verified. For this purpose more than 20 batch experiments have been performed.

In Fig. 15 the production times of 13 batch runs are indicated. The average separation difficulty is calculated for two values of α as the real α of the separation is unknown. The top vapour flow in the model is tuned to the one in the real column (2.7 mole/s). The following conclusions can be made.

- Experimental reproducibility. The experimental results show good reproducibility.
- Slope of relation. The results are plotted for two different relative volatilities. The relative volatility assumed in $\sigma_{dif,avg}$ influences the slope of the relation. The lines of $\alpha = 2.0$ slightly converge for higher values, whereas the lines of $\alpha = 2.15$ slightly diverge. The effective relative volatility appears to be approximately 2.10.
- Abscissa of relation. The lines have about the same slope as in theory but their abscissas differ clearly. The reason appears to be the behaviour of the initial production phase. During the first part of the production period the reflux ratio remains high since the column has not yet the correct purity profile. Also the temperature course shows considerable initial profile changes. The experimental lines depend on the start-up procedure. But it appears that if the distillation procedure is always strictly followed, then the production time can be estimated in a straightforward manner from the linear relation for the average separation difficulty.

9. Conclusions

The relative performance of the constant quality controller and the constant reflux controller compared to dynamic optimal control can be described with two variables: the difficulty of separation, σ_{dif} and the degree of exhaustion, σ_{exh}. Constant reflux control performs at $\sigma_{exh} < 60\%$ as good as dynamic optimal control, whereas constant quality control performs well when either σ_{dif} or σ_{exh} is low. The combination of these two specific measures gives a better understanding of the relative control performances than known up to now.

Batch distillation with slop recycling under constant quality control promises to offer a good alternative for dynamic optimal control. From the optimisation studies it appears that slop quantity and quality are always tuned in such a way that either σ_{dif} or σ_{exh} is low. During the product phase the degree of exhaustion, σ_{exh}, is kept low, whereas during the slop phase the degree of separation difficulty, σ_{dif}, is restricted. This applies for simple binary distillation, but also for ternary distillation, and binary distillation with considerable hold-up.

For a binary cut two degrees of freedom are available, namely slop quantity and quality. For a multiple batch distillation a third degree of freedom is added: the ratio of the more volatile and less volatile impurities in a product. This requirement links the binary distillations two by two. The slop amount can be determined by a relation in the slop indication measure, σ_{slop}, whereas slop quality can be fixed to a suitable value since sensitivity in this degree of freedom is small. The impurity ratio can be set equal to the ratio of relative volatilities between the less volatile (heavy) and the more volatile (light) impurity. This ratio can be corrected if the relative volatility of the less volatile component is very low.

Total batch time (time required for product and slop cuts together) which is important to compile a schedule, appears to have a linear relation with the average difficulty of the production runs, $\sigma_{dif,avg}$. The linear relation can be described by the abscissa and the slope. The slope appears to be independent of hold-ups. Also for ternary distillations the same slope was found. This relation only applies for batches with slop recycling. Apparently, for a certain column and product requirement the slop is tuned according to a fixed pattern described by the combination of exhaustion and difficulty of separation. For ternary distillation and for columns with large hold-up the required batch time for the same product amount is higher. This is caused by the higher difficulty of separation. The abscissa in the relation becomes larger. In practice, the two parameters, the slope and the abscissa, can be adapted easily to the actual column and product requirements. The relation has been experimentally verified and has proven to be applicable if a fixed distillation procedure is followed.

Nomenclature

A, B, C	components with decreasing volatility
B	bottom quantity (mole)
D	distillate quantity (mole)
$D_{\max}$	maximal D under a certain control policy
$D_{\text{potential}}$	theoretical maximal D ($=F\times x_F/x_D$)
F	feed quantity (mole)
L	reflux low (mole/s)
n	number of trays
T	production period (time)
V	vapour rate (mole/s)
x	– molar fraction of liquid; – binary: molar fraction of light component
y	molar fraction of vapour
α	relative volatility
$\varepsilon_{\text{column}}$	overall column efficiency
ε_V	mean Murphree vapour tray efficiency
σ_{dif}	degree of separation difficulty, Eq. (4)
$\sigma_{\text{dif,avg}}$	average σ_{dif} during product run, Eq. (8)
σ_{exh}	degree of exhaustion, Eq. (5)
σ_{slop}	slop indication measure, Eq. (7)

Indices

0	at start of run (t=0)
A, B, C	components with decreasing volatility
B	bottom
D	distillate
F	feed
AB, BC	separation A–B, B–C
CQ	constant quality control
CR	constant reflux control
DO	dynamic optimal control
PC	product cut
SC	slop quantity (mole)
set	set value

Appendix A

Column model

The following assumptions apply for the column model:

- The distillation dynamics are described by the column exhaustion. In some simulations tray hold-up has been added.
- The relatively fast fluid dynamics are neglected: $L_{i+1}=L_i$.
- The pressure drop is neglected since the drop is small compared to the absolute pressure.
- The distillation operates adiabatically: $V_{i+1}= V_i$.
- The relative volatility is constant and independent of the composition.

Since the tray and accumulator mass is constant, the mass leaving the column seems to be withdrawn from the bottom directly. The production and reflux are proportional to the vapour flow. For the bottom molar mass and composition applies:

$$\frac{\mathrm{d}B}{\mathrm{d}t} = -\frac{\mathrm{d}D}{\mathrm{d}t} = -V + L \tag{A1}$$

$$B\frac{\mathrm{d}x_0}{\mathrm{d}t} = \left[\frac{\mathrm{d}(Bx_0)}{\mathrm{d}t} - x_0\frac{\mathrm{d}B}{\mathrm{d}t}\right] = -V(y_0 - x_0) + L(x_1 - x_0) \tag{A2}$$

Product vessel increase directly follows the bottom exhaustion

$$D\frac{\mathrm{d}x_D}{\mathrm{d}t} = (V - L) \times (y_n - x_D) \tag{A3}$$

Tray liquid composition can be calculated from the operating line:

$$x_i = \frac{V}{L}y_{i-1} - \frac{V - L}{L}y_n \tag{A4}$$

Vapour composition is determined by the vapour–liquid equilibrium with constant volatility and the Murphree tray efficiency.

$$y_i^{\mathrm{e}} = \frac{\alpha x_i}{(\alpha - 1).x_i + 1} \quad \text{with} \quad \varepsilon_V = \frac{y_i - y_{i-1}}{y_i^{\mathrm{e}} - y_{i-1}} \tag{A5}$$

In the case of ternary distillation a few additions are required. Ternary distillation is described by two operating lines, two equilibrium relations, and two tray efficiency relations. Relative volatility is related to the heavy component C. Thus: $\alpha_B = \alpha_{BC}$, and $\alpha_A = \alpha_{AC} = \alpha_{AB}\alpha_{BC}$. In the simulations always $\varepsilon_{V,A}$ is set equal to $\varepsilon_{V,B}$. The tray compositions can only be solved iteratively.

References

[1] E. Quintero-Marmol, W.L. Luyben, Multicomponent batch distillation 2. Comparison of alternative slop handling operating strategies, Ind. Eng. Chem. Res. 29 (1991) 1915–1921.

[2] D.W.T. Rippin, Simulation of single- and multi-product batch chemical plants for optimal design and operation, Computers Chem. Eng. 7(3) (1983) 137–156.

[3] E.R. Robinson, Optimum reflux policies for batch distillation, Chem. Process Eng. 5(71) (1971) 47–55.

[4] L.H.J. Kerkhof, H.J.M. Vissers, On the profit of optimum control of batch distillation, Chem. Eng. Sci. 33 (1978) 961–970.

[5] E.R. Robinson, The optimal control of an industrial batch distillation column, Chem. Eng. Sci. 25 (1970) 921–928.

[6] T.T. Hansen, S.B. Jørgensen, Optimal control of binary distillation in tray or packed columns, Chem. Eng. J. 33 (1986) 151–155.

[7] S. Farhat, M. Czernicki, L. Pibouleau, S. Dominech, Optimization of multiple-fraction batch distillation by non-linear programming, AIChE J. 36(9) (1990) 1349–1360.

[8] J.R. Bosley, Jr., T.F. Edgar, Application, of non-linear model predictive control to optimal batch distillation. IFAC DYCORD+ 1992, MD, USA, (1992), 303–308.

[9] U.M. Diwekar, R.K. Malik, K.P. Madhavan, Optimal reflux rate policy determination for multicomponent batch distillation columns, Computers Chem. Eng. 11(6) (1987) 629–637.

[10] U.M. Diwekar, Unified approach to solving optimal design-control problems in batch distillation, AIChE J. 38(10) (1995) 1551–1563.

[11] D.N. Mayur, R.A. May, R. Jackson, The time-optimal problem in binary batch distillation with a recycled waste-cut, Chem. Eng. J. 1 (1970) 15–21.

[12] D.N. Mayur, R. Jackson, Time-optimal problems in batch distillation for multicomponent mixtures and columns with holdup, Chem. Eng. J. 2 (1971) 150–162.

[13] F.M. Christensen, S.B. Jørgensen, Optimal control of binary distillation with recycled waste cut, Chem. Eng. J. 34 (1987) 57–64.

[14] W.L. Luyben, Multicomponent batch distillation. 1 Ternary systems with slop recycle, Ind. Eng. Chem. Res. 27 (1988) 642–647.

[15] I.M. Mujtaba, S. Macchietto, An optimal recycle policy for multicomponent batch distillation, Computers Chem. Eng. 16 (1992) S273–280.

[16] I.M. Mujtaba, S. Macchietto, Optimal operation of multi-component batch distillation – multi-period formulation and solution, Computers Chem. Eng. 17(12) (1993) 1191–1207.

[17] L. Bonny, S. Dominech, P. Floquet, L. Pibouleau, Strategies for slop cut recycling in multi-component batch distillation, Chem. Eng. Processing 33 (1994) 23–31.

[18] B.H.L. Betlem, Hierarchical control of batch distillation, Ph.D. Thesis, University of Twente, Enschede, Netherlands, 1996.