Optimization of Multiple-Fraction Batch Distillation with Recycled Waste Cuts

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A strategy for the optimal operation of multiple-fraction batch distillation processes with waste cut recycling is presented. The processes are modeled by differential algebraic equations formulated in the gPROMS modeling language. A cost function based on economic criteria is developed. Recycling of the waste cuts is formulated as a cyclically repeated batch distillation process, where the waste cuts are recycled at intermediate points in time. The resulting multipoint boundary value optimization problem is efficiently solved by using the dynamic optimization package MUSCOD-II. A comparison of optimization results for the separation of a ternary mixture shows that considerable gains in profits can be obtained by a complete recycling of the waste cuts. Computation times are in the range of minutes on a standard workstation.

Introduction

Batch distillation processes are a rewarding field for nonlinear optimization techniques: due to considerable changes of the operating conditions over the process time, the inherently nonlinear model equations cannot be simplified by linearization, and, due to high operating costs and thin profit margins, even small performance gains are worth considerable effort.

The direct multiple shooting method (Bock and Plitt, 1984), combined with a *partially reduced SQP technique* (Leineweber, 1999), recently implemented in the dynamic optimization package MUSCOD-II, is well suited to efficiently solve optimization problems of this kind. Among the features of MUSCOD-II are the possibility to provide the DAE model equations as generic C or Fortran-Code or in the gPROMS modeling language (Leineweber et al., 2002), to make use of efficient state-of-the-art DAE solvers (such as DDASAC by Caracotsios and Stewart (1985) or DAESOL by Bauer et al. (1999)), or to employ an existing parallelization in the portable MPI standard, in time critical cases. An example of a quite intricate application problem with semi-periodic boundary conditions is presented in the following.

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Waste cut recycling problems were treated, for example, by Mayur et al. (1970) and Christensen and Jorgensen (1987) for binary batch distillation, where it was possible to find time optimal reflux policies in the framework of Pontryagin's maximum principle. Luyben (1988, 1990) treats a ternary mixture, but does not solve an optimal control problem. In contrast to this, we optimize stage durations, recycling ratios and controls simultaneously for a ternary mixture, using a cost function that incorporates energy costs, product prices, and possible waste cut disposal costs.

Batch Distillation Process

The multiple-fraction batch distillation process has as its goal the separation of a mixture of n components (referred to as feed in the following) into different fractions of prespecified purity levels.

In this study we treat the complete separation of a ternary mixture, which is accomplished by four consecutive distillation phases: the lightest component is separated in a first production cut, and remaining traces of it are removed in a following *waste cut* (also referred to as (*slop cut*), which helps to attain the desired purity of the second lightest fraction in the following second production cut. The purity of the third

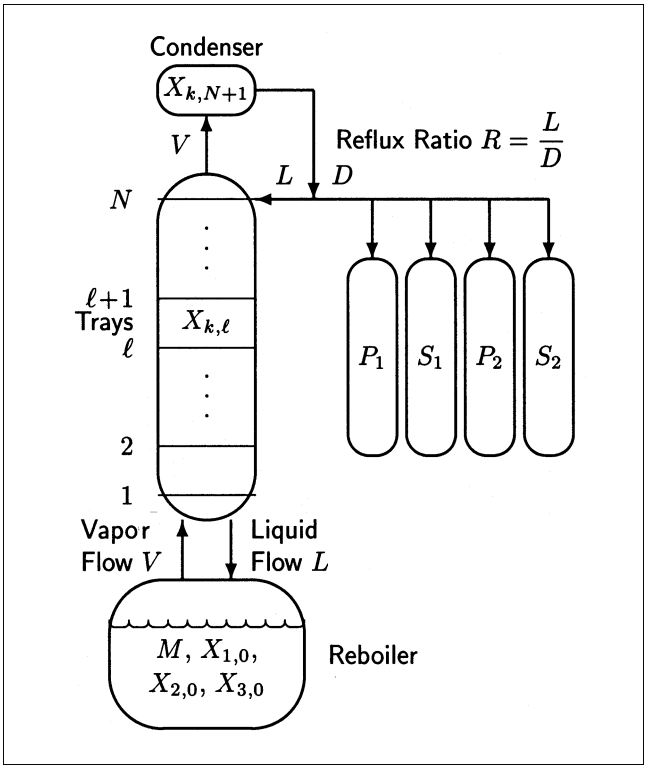

Figure 1. Ternary batch distillation with two production cuts P_1, P_2 and two waste cuts S_1, S_2 in a column with N ideal trays.

and heaviest fraction is achieved by a last waste cut that removes remaining impurities from the bottoms product.

The process is controlled by the *reflux ratio* R, which can be varied over time. The distillation column and the process under investigation is shown in Figure 1.

DAE-Model of the Batch Distillation Column

The mathematical model of the distillation column, developed, for example, by Farhat et al. (1990), is based on the following simplifying assumptions:

- Ideal trays
- No liquid holdup
- No pressure drop
- Total condenser
- Constant molar overflow for vapor V and liquid L
- Tray phase equilibria as for ideal mixture, with partial pressures determined by the Antoine-equation.

We assume that the heating power is kept constant, so that the vapor flow V is not a control, but a fixed parameter. Using the reflux ratio R, L and D can directly be eliminated

$$L = V\frac{R}{R+1} \text{ and } D = V\frac{1}{R+1} \tag{1}$$

The concentrations in the reboiler are denoted by $X_{k,0}$ and those in the condenser by $X_{k,N+1}$, analogously to the tray concentrations ($x_{k,\ell}$).

The only non-neglected mass in the system is the molar reboiler content M. During the distillation process, M is reduced by the distillate flow D

$$\frac{dM}{dt} = -D = \frac{-V}{R+1} \tag{2a}$$

The mass conservation of the different components k requires analogously

$$\frac{d(MX_{k,0})}{dt} = -DX_{k,N+1}$$

which is equivalent to

$$\frac{d(X_{k,0})}{dt} = \frac{V}{M(R+1)}(X_{k,0} - X_{k,N+1}) \tag{2b}$$

for $k = 1, 2$. The conservation of the third component is implicitly given by Eqs. 2a and 2b. Therefore, the three dynamic equations for the differential variables M, $X_{1,0}$, and $X_{2,0}$ are specified.

As algebraic equations, we first require the componentwise mass conservation in the column section above the ($\ell+1$)st tray

$$VK_k(T_\ell)X_{k,\ell} - LX_{k,\ell+1} - DX_{k,N+1} = 0$$

or equivalently

$$K_k(T_\ell)X_{k,\ell} - \frac{R}{R+1}X_{k,\ell+1} - \frac{1}{R+1}X_{k,N+1} = 0 \tag{2c}$$

for $k = 1,2$ $\ell = 0, 1, \ldots, N$. The first term corresponds to the vapor flow entering into the ($\ell+1$)st tray, the second to the outflowing liquid, and the third to the distillate flow D. The vapor concentrations on the ℓth tray are calculated as the product of the equilibrium values $K_k(T_\ell)$ with the liquid concentration $X_{k,\ell}$.

The concentration of the third component is determined by the closing condition

$$1 - \sum_{k=1}^{3} X_{k,\ell} = 0, \qquad \ell = 0, 1, \ldots, N+1 \tag{2d}$$

The tray temperature T_ℓ is implicitly defined by the closing condition for the vapor concentrations

$$1 - \sum_{k=1}^{3} K_k(T_\ell)X_{k,\ell} = 0, \ \ell = 0, 1, \ldots, N \tag{2e}$$

Assuming an ideal mixture, the equilibrium values $K_k(T_\ell)$ are

Table 1. Material Properties for Batch Distillation*

k	Boiling Point (°C)	Antoine Coefficients		
		A_k	B_k	C_k
1	184.4	7.63846	1976.3	231.0
2	245.0	7.96718	2502.2	247.0
3	272.5	8.65385	3149.1	273.0

*Components 1, 3 and 7 from Domenech and Enjalbert (1981) and Farhat et al. (1990).

determined according to Raoult's law

$$K_k(T) = \frac{P_k^s(T)}{P}, \quad k = 1, 2, 3 \tag{2f}$$

Here, P is the total pressure assumed to be constant over the whole column, and $P_k^s(T)$ are the partial pressures of the undiluted components: they are determined by the Antoine equation for $k = 1, 2, 3$

$$P_k^s(K) = \exp_{10}\left(A_k - \frac{B_k}{T + C_k}\right) \tag{2g}$$

The Antoine coefficients used in the presented example are given in Table 1. The total number of algebraic variables ($X_{3,0}$, $X_{k,\ell}$ for $k = 1, 2, 3$ and $\ell = 1, \ldots, N+1$, as well as T_ℓ for $\ell = 0, \ldots, N$) in the given formulation is $4N + 5$. By resolving Eq. 2d, the concentrations $X_{3,\ell}$ for $\ell = 1, 2 \ldots, N$ can be eliminated directly, so that only $3N + 5$ algebraic states remain. In the computations presented in this article we have used a model with $N = 5$ trays, formulated in the gPROMS modeling language (Process Systems Enterprise, 2000).

The described model, with holdups neglected on trays and in the reflux drum, is similar to models considered by Diwekar et al. (1987) or Logsdon et al. (1990). It has to be noted, however, that the inclusion of holdups in the model may lead to quite different optimal profiles, as demonstrated, for example, by Logsdon and Biegler (1993) and Mujtaba and Macchietto (1998). Other approaches towards batch distillation modeling and optimization have been developed by Diwekar (1995) and by Mujtaba and Macchietto (1992, 1996).

Formulation of the Optimization Problem

The question examined in this article is whether it is profitable to recycle the waste cuts instead of removing them from the distillation process. For the separation of an amount of feedstock considerably larger than a single batch, carried out by a sequence of identical batches, this could be achieved by adding the waste material of one batch to the feed of the following one. Although this reduces the amount of *fresh* feedstock which can be processed in a single batch (due to limited reboiler holdup), and, thus, results in a longer overall distillation time T for a given total amount of feedstock, the product outputs P_1, P_2 and P_3 are increased and disposal of the slop cuts S_1 and S_2 becomes unnecessary.

If the product prices are specified by constants p_i, and the costs for distillation time/energy consumption by c_{energy} and for slop-cut disposal by s_j, the profit P for the whole process is given by

$$P = \sum_{i=1}^{3} p_i P_i - \sum_{j=1}^{2} s_j S_j' - c_{\text{energy}} T \tag{3}$$

where S_j' are the amounts of slop-cut material that are *not* recycled. The costs for the feed purchase are constant for a fixed amount of feedstock and are left out of consideration in the problem formulation.

To treat the recycling problem, we consider the limiting case of a total amount of feed that is considerably larger than the reboiler holdup; this would result in a large number of consecutive, nearly identical batches: here, one batch produces the amount of slop-cut material that is available for recycling in the following batch. If we assume that all batches are identical, we need to treat only *one single* batch which produces exactly the same slop-cut material as its output which was previously given to it as an input for recycling. This quasi-periodic process formulation leads to a coupled multipoint boundary value problem.

The first slopcut is added at the beginning of the first production cut, as it contains mainly components 1 and 2, while the second slop cut is recycled in the second production cut, because it mainly consists of components 2 and 3.

To allow for *partial* recycling of the slop cuts, we have introduced the recycling ratios R_1, R_2 as *free* optimization parameters. Thus, the nonrecycled parts of the slop cuts become

$$S_j' = (1 - R_j) S_j, \qquad j = 1, 2 \tag{4}$$

For the determination of the total distillation time, we proceed as follows: instead of determining the increased number of batches due to the slop-cut recycling, we artificially increase the initial reboiler filling of a single batch by the amount of the recycled material from slop cut S_1—this leaves the total amount of *fresh* fixed, but increases the time for one batch by exactly the same factor, by which the amount of batches should have been increased. This is due to the fact that the differential Eqs. 2a and 2b are invariant with respect to the simultaneous rescaling of time t and reboiler content M. Note also that the vapor flow V enters all differential equations linearly and only leads to a rescaling of the time axis. Therefore, by varying V as a control over time (instead of keeping it constant), no additional gains would be produced in the considered cost model.

If we neglect the cost of the preparation time needed between two batches (during which no energy costs occur), we can restrict the objective formulation to material and energy costs of one single batch only.

We note that experimentation with alternate problem formulations, such as the fixing of production and optimization of raw material, are easy to accomplish in the framework of MUSCOD-II. For the sake of simplicity, however, we restrict ourselves to one single formulation throughout this article.

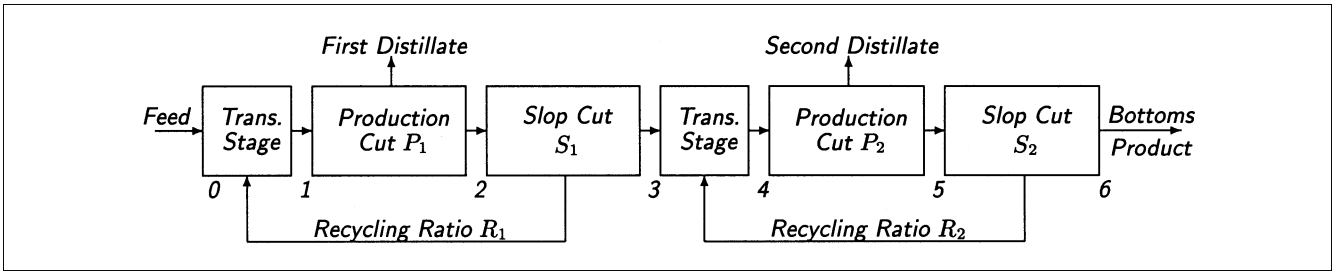

Figure 2. Quasi-periodic ternary batch distillation with waste cut recycling.

Coupled Multipoint Boundary Value Constraints of the Recycling Problem

The partial recycling of the slop cuts results in interior point constraints on the value of the reboiler holdup B, which is illustrated in Figure 2. By conceptionally introducing intermediate *transition stages* to describe the addition of the slop cuts, we can formulate these constraints as a linear coupling between the value of M at the interior points 0, 1, …, 6 in the following way

$$M|_1 - M|_0 = R_1 S_1 = R_1(M|_2 - M|_3) \tag{5a}$$

$$M|_4 - M|_3 = R_2 S_2 = R_2(M|_5 - M|_6) \tag{5b}$$

Analogously, the transition equations for the two other differential variables, the reboiler concentrations $X_{k,0}$ $(k = 1, 2)$, are formulated by using the conservation of the component quantities $X_{k,0}M$

$$X_{k,0}M|_1 - X_{k,0}M|_0 = R_2\left(X_{k,0}M|_2 - X_{k,0}M|_3\right), \quad k = 1, 2 \tag{6a}$$

$$X_{k,0}M|_4 - X_{k,0}M|_3 = R_2\left(X_{k,0}M|_5 - X_{k,0}M|_6\right), \quad k = 1, 2 \tag{6b}$$

Altogether, we obtain six coupled interior point constraints that determine the jumps of the three differential states in the two transition stages.

The purity requirements for the three product fractions are imposed by additional interior point constraints

$$\frac{X_{1,0}M|_1 - X_{1,0}M|_2}{M|_1 - M|_2} \geq X_{P_1} \tag{7a}$$

$$\frac{X_{2,0}M|_4 - X_{2,0}M|_5}{M|_4 - M|_5} \geq X_{P_2} \tag{7b}$$

$$X_{3,0}|_6 \geq X_{P_3} \tag{7c}$$

Here, $X_{P_1} = 98\%$, $X_{P_2} = 96\%$, and $X_{P_3} = 99\%$ are the required minimum purities of the main component in the product fractions.

Optimization Results

Two different kinds of optimizations have been carried out for comparison: a first one without any recycling of the slop cuts, that is, with recycling ratios R_1, R_2 fixed to be zero, and a second one following the description given above, with full freedom to choose the recycling ratios between 0 and 1.

In the example presented, the assumptions for costs and prices are made as careful as possible not to encourage the recycling of slop cuts: no costs at all are assigned to slop-cut disposal and the prices for the products are chosen so low that, in the nonrecycling case, the gains are only marginally higher than the energy consumption costs:

- Slop-cut disposal costs: $s_j = 0.00$ for $j = 1, 2$;
- Product prices: $p_i = 4.50$ for $i = 1, 2, 3$; and
- Time/energy consumption costs: $c_{\text{energy}} = 1.0$

If the feedstock purchase costs were taken into consideration, the product prices should be even higher to retain marginal profitability of the process.

The feed is assumed to contain all three components in equal amounts, that is, $X_{k,0}|_0 = 0.33$ for $k = 1, 2$. The starting mass of feed is set to $M|_0 = 1$. The parameterization of the control—the reflux ratio $R(t)$—is chosen to be continuously piecewise linear on the four model stages. The chosen multiple shooting parameterization can be seen in Figures 3 and 4—each multiple shooting interval corresponds to one free control parameter. In addition to the controls, the stage durations are also free for optimization.

The DAE solution and derivatives are calculated by DAESOL (Bauer et al., 1999; Bauer, 1999), an integrator based on the backward-differentiation-formulae (BDF).

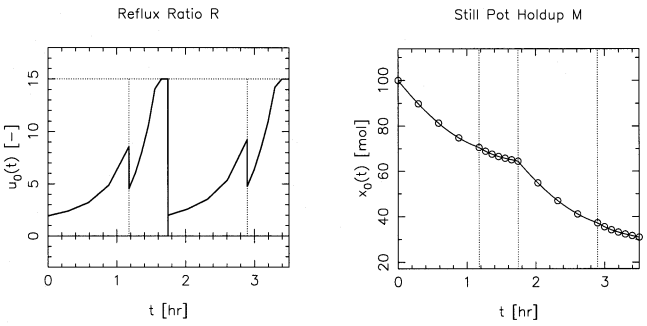

Figure 3. Solution profile for the batch distillation *without* slop-cut recycling.

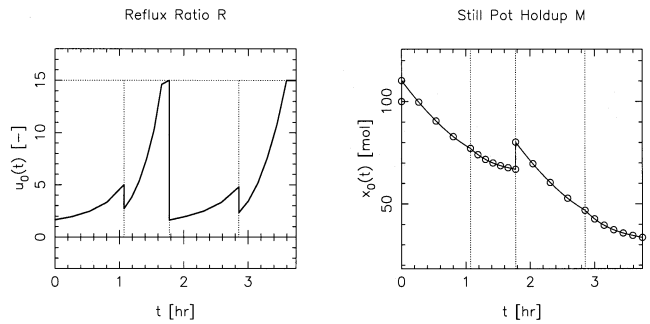

Figure 4. Solution profile for the batch distillation *with* slop-cut recycling.

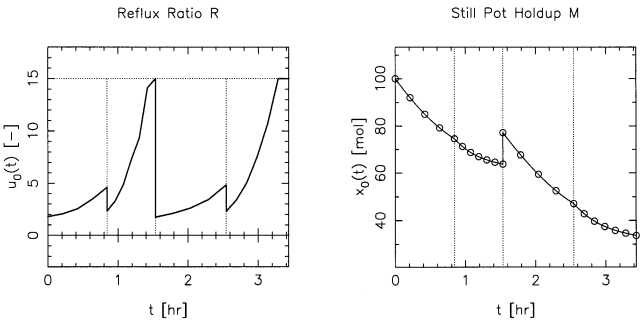

Figure 5. Solution profile for a lower product price $p_1 = 2.00$ with optimal recycling ratios $R_1 = 0$ and $R_2 = 1$.

Optimization without waste cut recycling

With the values given above, the optimization for the nonrecycling case yielded process duration (energy) costs of 3.51 units, and a product sale income of 3.96 units, that is, 0.45 units of profits.

The slop-cut outputs are small, with relatively short durations and high reflux ratios, as these outputs are lost for the process (cf. Figure 3).

Optimization with waste cut recycling

When a slop-cut recycling was allowed, the optimization result was an increased total time with an energy cost of 3.74 units and product yields of 4.50 units. The net profits of 0.76 units are increased by 69% compared to the no-recycling value of 0.45.

The reflux ratio is generally smaller than before, as the purity constraints are satisfied by taking bigger slop-cuts that are no longer lost for the process (cf. Figure 4).

As the optimizer had full freedom to choose the recycling ratios R_1, R_2 between 0 and 1 and was initialized with the values 0, but found at the end of the optimization that for both the value 1 is optimal, we can practically exclude the possibility that partial recycling would have been more profitable. Tests with different initial guesses (such as with the optimal solution of the nonrecycling case) confirm this claim.

The computation times for both optimization problems are well below 3 min on a Sun ULTRA 10 workstation.

Further optimization studies

We have performed further optimization studies with varying cost coefficients. In all scenarios a slop-cut recycling was allowed, but the optimizer was always initialized with a feasible trajectory *without* slop-cut recycling, that is, $R_1 = R_2 = 0$.

Experiments with positive slop-cut disposal costs $s_j > 0$ (but unchanged product prices and energy costs) confirm the expectation that a complete slop-cut recycling is even more advantageous in this case, and even greater relative gains are obtained. The optimal solution profiles with slop-cut recycling are identical to the one shown in Figure 4.

When one product price p_1 was reduced to lower values, starting from the nominal value $p_1 = 4.50$ and stepwise decreased by 0.25, recycling of the first slop cut becomes unprofitable for $p_1 < 2.25$, and no recycling of the first slop cut is recommended by the optimizer. These results were confirmed by checking with different initial guesses.

The optimal profile for $p_1 = 2.00 < 2.25$, is shown in Figure 5, with the optimal recycling ratio values $R_1 = 0$ and $R_2 = 1$.

Conclusions

Ternary batch distillation with recycled waste cuts is considered as an example for dynamic optimization with semi-periodic boundary conditions.

The problem is formulated as a multistage optimization problem with an objective function that takes energy costs and product yields into account. The DAE model equations are provided in the gPROMS modeling language. The recycling of the waste cuts is modeled by transition stages that allow for jumps in the differential variables. The aim to find a stationary cycle results in semi-periodic multipoint constraints. The problem formulation presented can easily be extended to multiple-fraction batch distillation processes with more than three components.

The problem is solved by the use of the dynamic optimization package MUSCOD-II, which is designed to efficiently treat multistage problems with multipoint constraints.

For the formulated example problem, it is shown that considerable gains can be obtained by an optimized complete recycling of the waste cuts when compared to the optimal solution *without* recycling of the waste cuts.

The ease of implementation and favourable computation times in the range of minutes make MUSCOD-II an advantageous tool for investigation of different scenarios, sensitivity analysis, and for quick re-optimization as prices change.

Moreover, an extension of MUSCOD-II to real-time optimization problems and nonlinear model predictive control has been developed (Diehl et al., 2002) and was successfully tested on a medium-scale continuous distillation column located at the University of Stuttgart (Diehl et al., 2001a; Diehl, 2002).

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