ON THE SIMULTANEOUS OPTIMAL DESIGN AND OPERATION OF BATCH DISTILLATION COLUMNS

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We explore a simultaneous optimisation strategy for the design and operation of batch distillation columns. A number of researchers have dealt with the optimal operation (e.g. optimal reflux policy) of these systems. These studies are limited to the optimisation of column operations keeping the column design fixed

By applying high order implicit methods (such as collocation on finite elements), one obtains a stable and highly accurate algebraic representation of the differential equations, which can be written as constraints within the optimisation problem. With this approach, both solution and optimisation of the batch column model occur simultaneously. This simultaneous approach allows for problem formulations that deal with the integration of process design and operation, since optimal design parameters and operating policies are optimised together.

An objective function is formulated that maximises the net profit, defined as the value of the product minus the capital costs for the batch distillation unit. The short-cut model (Diwekar, 1988) based upon Fenske's, Gilliland's, and Underwood's correlations for column design is used to allow the number of trays to be a continuous variable, within the nonlinear program. Here we find the level of approximation for the short-cut model is valid for these preliminary design cost studies.

This approach is demonstrated on a number of applications including the design and operation of a column for a single feed composition and the multiperiod problem for the design of a single column to separate different feed charges that have different compositions and components. This approach thus allows the consideration of optimal operation at the design stage and includes the consideration of multiple services in the design.

1. INTRODUCTION

Due to the increasing importance of speciality chemicals, the optimal design and operation of process equipment for batch processes is of interest, especially for an operation that uses expensive raw materials. The determination of an optimal control policy for an existing unit operation has been investigated over the years for batch distillation. However, the simultaneous design and operation of the batch columns for an economic objective function has received little attention in the literature. Diwekar *et al.* (1989) investigated the design of the column for an economic objective function that considered capital costs. The optimal control profile was also found for the fixed column design by Diwekar *et al.* (1987). In this paper, we explore a strategy for the simultaneous design and operation of batch distillation columns. We want to ensure that at the preliminary design stage, the column will be able to accomplish the desired separation for different components and mixtures in an economic manner so that the operation is profitable. We include in our profit function the capital costs for the column and utility costs, and balance these costs against the value of the product. Once the column parameters have been determined, then a rigorous model can be used to verify the control profile optimization. Simultaneous design and operation is useful especially for multipurpose columns, where the optimal design is required to handle more than one operation.

A number of researchers have investigated the optimal operation of the column once the column design is known. The classic cases considered are:

1. Maximum Distillate Problem—Maximise the amount of distillate of a specified concentration for a specified batch time.

2. Minimum Time Problem—Minimise the batch time needed to produce a prescribed amount of distillate of a specified concentration.

3. Maximum Profit Problem—Maximise a profit function for a specified concentration of distillate.

As mentioned above, Diweker *et al.* (1989) investigated the design of the column for an economic objective function that considered capital costs; in a previous study they also solved the Maximum Distillate Problem (Diwekar *et al.*, 1987). In both studies a shortcut approach using Fenske's, Gilliland's and Underwood's correlations was used for the modelling in order to make the number of plates a continuous variable. Mass and energy balances were written in their differential form for dynamic modelling. Pontryagin's maximum principle was used to solve the Maximum Distillate Problem for the short-cut model and the results were equivalent to the plate to plate calculations reported in the literature. Columns handling binary and multicomponent systems were optimised using this approach.

Kerkhof and Vissers (1978) combined the Maximum Distillate Problem and the Minimum Time Problem to form a profit function in which they maximised the profit on an hourly basis for a fixed design. The systems studied were binary distillation columns using a plate to

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2. MATHEMATICAL MODEL

Here we are confronted either with a mixed integer nonlinear programming (MINLP) problem to determine the number of trays rigorously, or a carefully chosen model simplification so that N can be treated as a continuous parameter. Viswanathan and Grossmann (1990) used a MINLP approach for steady state column design, but this can be potentially expensive for batch columns with dynamics. Diwekar (1988) has shown the effectiveness of using a short-cut model for preliminary design purposes. We briefly review the short-cut formulation and show how a mathematical programming approach can be used to solve the optimal control problem while simultaneously designing the column in order to achieve a maximum profit. The short-cut methods are based on constant relative volatilities of the components. In addition, Fenske's equation, Underwoods's equations, and Gilliland's correlation are used to determine the optimal values for the reflux ratio profile and the number of trays for a specified purity constraint to maximise the profit function. We rewrite Fenske's equation in the form of Hengstebeck-Geddes' equilibrium equation. The following problem formulation results:

Maximise

$R_t(t), N, V_f$

$$\psi = \frac{DP_r - B_0 C_0}{t_f + T_s} - \frac{K_1 V^{0.5} N^{0.8} + K_2 V^{0.65} + K_3 V}{\text{Hrs.}} \tag{3}$$

s.t.

Mass balances

$$\frac{dS}{dt} = -\frac{V}{R_t + 1}, \quad S = \text{overall mass} = B_0 - D(t) \tag{4}$$

$$\frac{dx_b^1}{dt} = -\frac{V[x_b^1 - x_d^1]}{[R_t + 1]S} \quad x_b^1 = \text{key component} \tag{5}$$

$$\frac{dx_b^j}{dt} = -\frac{V[x_b^j - x_d^j]}{[R_t + 1]S} \quad j = 2, \ldots, n - \text{no. of components} \tag{6}$$

Underwood's Correlation

$$0 = \sum_{j=1}^{n} \frac{\alpha_j x_b^j}{\alpha_j - \varphi}; \quad R_{min} + 1 = \sum_{j=1}^{n} \frac{\alpha_j x_d^j}{\alpha_j - \varphi} \tag{7}$$

Gilliland's Correlation

$$y = 1 - \exp\left[\frac{(1 + 54.4X)(X - 1)}{(11 + 117.2X)\sqrt{X}}\right] \tag{8}$$

$$X = \frac{(R_t - R_{min})}{(R_t + 1)} \quad \text{and} \quad y = \frac{(N - N_{min})}{(N + 1)}$$

Hengstebeck-Geddes' equilibrium equation

$$\frac{x_d^j}{x_b^j} = \left[\frac{\alpha_j}{\alpha_1}\right]^{N_{min}} \frac{x_d^1}{x_b^1}, \quad j = 1, \ldots, n \tag{9}$$

Summation equations

$$\sum_{j=1}^{n} x_d^j = 1.0 \quad \sum_{j=1}^{n} x_b^j = 1.0 \tag{10}$$

Purity constraint

$$x^1_{d,\text{lower bound}} \le x_d^* = \frac{\int_0^{t_f} \frac{x_d^1 V}{R_t + 1} dt}{\int_0^{t_f} \frac{V}{R_t + 1} dt} \le 1.0 \tag{11}$$

where

R_t = reflux ratio
x_d^1 = composition of distillate (key component)
x_d^j = compositions of distillate (other than key)
x_b^1 = composition in reboiler (key component)
x_b^j = composition of reboiler (other than key)
α_j = relative volatilities
φ = Underwood's roots
x_d^* = final average composition of product in distillate receiver.

If constant molal overflow is not an adequate assumption, then the differential enthalpy balances can be added as required. Also, this short-cut model can be modified to include the dynamic relations that describe the effect of holdup in the condenser system for simulation purposes once the number of trays has been determined.

The shortcut model itself only needs the feed composition in order to determine the column design for a desired purity specification. For the simultaneous approach we find optimal values for $R_t(t)$, N, V and the optimal batch time, t_f. This optimisation problem allows us to make a simplification with regard to vapour boilup rate. Vapour boilup rate for operation $V(t)$ is constrained by a maximum rate, V_{design}, determined by the column diameter. However, one can show with an intuitive (as well as a variational) argument that $V(t) \doteq V_{design}$ if the optimal operation is profitable and (3) is our objective function. Consequently, boilup rate can be described simply by the variable V for this problem.

Note that the overall mass balance gives a direct relationship between the final batch time and the amount of distillate produced. For convenience, in our shortcut model we assume V constant and have

$$\frac{dS}{dt} = -\frac{V}{R_t + 1} = \frac{-dD}{dt} \tag{12}$$

By introducing a dimensionless time,

$$\bar{t} = \frac{t}{t_f}$$

we normalise the final batch time between 0 and 1 to get:

$$t_f = \frac{D}{VR_{avg}}, \quad R_{avg} = \int_0^1 \frac{d\bar{t}}{R_t + 1} \tag{NT}$$

This expression can be substituted for t_f in the objective function, if required. Note that the differential equations are modified to include the final batch time, t_f. This greatly simplifies the formulation by allowing t_f to become an additional parameter in the optimisation. Also, it can be shown for this problem that the optimum value for V lies at its upper bound if setup time T_s is bounded by a multiple of t_f (Appendix B). While this assumption may not always be realistic, the upper bound on V is frequently dictated by exogenous considerations such as maximum product demand or other equipment

Problem (NLP1) can now be solved by any large scale nonlinear programming solver. For this study we used the GAMS modelling system to set up (NLP1) and obtained solutions with the RND/SQP strategy of Vasantharajan and Biegler (1988). It should be noted that (NLP1) can also be simplified if none of the profile inequality constraints are active. Here we take advantage of Pontryagins's maximum principle to yield a smaller optimisation problem.

Application of Maximum Principle to NLP1

We first consider the Maximum Profit objective given by equation (1) and note that only D (amount of distillate) is directly influenced by R_t. Also for fixed N, V, and t_f we see that the Maximum Profit Problem merely becomes the Maximum Distillate Problem and the purity constraint is given by equation (11) or equivalently by:

$$\frac{x_{feed} B_0 - x_d^1(t_f) S(t_f)}{B_0 - S(t_f)} = \text{purity} \quad (19)$$

For given values of N, V, and t_f one can now derive the relations for optimal reflux policy through the variational conditions for the shortcut model (3). First, we form the Hamiltonian using differential equations (4) and (5):

$$H = \lambda_1 \frac{-V}{[R_t + 1]} + \lambda_2 \frac{V[x_b^1 - x_d^1]}{[R_t + 1]S} \quad (20)$$

and the adjoint equations,

$$\frac{d\lambda_1}{dt} = \frac{\lambda_2 V(x_b^1 - x_d^1)}{(R_t + 1)S^2} \quad (21)$$

$$\frac{d\lambda_2}{dt} = \frac{-\lambda_2 V\left(1 - \frac{\partial x_d^1}{\partial x_b^1}\right)}{(R_t + 1)S} \quad (22)$$

with final conditions

$$\lambda_1(t_f) = \frac{P_r}{t_f + T_s} + \frac{\mu_f B_0[x_b^1(t_f) - x_{feed}]}{[B_0 - S(t_f)]^2} \quad (23)$$

$$\lambda_2(t_f) = \frac{\mu_f\{S(t_f)\}}{[B_0 - S(t_f)]} \quad (24)$$

Here μ_f is the Lagrange multiplier for the purity constraint (19).

Now the two adjoints λ_1, λ_2 can be combined into one variable given by $\lambda = \lambda_2/\lambda_1$ and the adjoint equations can be combined to:

$$\frac{d\lambda}{dt} = -\frac{\lambda V\left(1 - \frac{\partial x_d^1}{\partial x_b^1}\right)}{(R_t + 1)S} - \frac{\lambda^2 V(x_b^1 - x_d^1)}{(R_t + 1)S^2} \quad (25)$$

with

$$\lambda(t_f) = f[\mu_f, S(t_f), x_b^1(t_f)] \quad (26)$$

The optimality condition on the reflux policy leads to

$$\frac{\partial H}{\partial R_t} = 0 \quad (27)$$

which implies

$$R_t = \frac{S - \lambda(x_b^1 - x_d^1)}{\lambda \frac{\partial x_d^1}{\partial R_t}} - 1 \quad (28)$$

Here $\partial x_d^1/\partial x_b^1$ and $\partial x_d^1/\partial R_t$ are derived from the short-cut model. Note that only the key component still compositions are expressed time explicitly. For the other components time implicit equations are useful.

These constraints are added to the nonlinear program so that the differential equality constraints to be solved are

$$\begin{aligned} \dot{S} &= f_1(S, x_b^1, x_d, R_t) \\ \dot{x}_b^1 &= f_2(S, x_b^1, x_d, R_t) \\ \dot{\lambda} &= f_3(S, x_b^1, \lambda, x_d, R_t) \\ R_t &= f_4(S, x_b^1, \lambda, x^d) \end{aligned} \quad (29)$$

We now apply orthogonal collocation to these equations and note that initial conditions for x_b^1 and S can be determined in the same way as with (NLP1). The final condition for λ is determined by adjusting μ_f until the purity constraint is met. We note that instead of specifying μ_f we simply leave the final condition for λ as a degree of freedom that is determined by direct enforcement of the purity constraint in the problem. The resulting NLP formulation now becomes:

$$\begin{aligned} &\max_{N, V, t_f} \quad \Psi \quad \text{(NLP2)} \\ \text{s.t.} \quad &\dot{S} = f_1(S, x_b^1, x_d, R_t) \\ &\dot{x}_b^1 = f_2(S, x_b^1, x_d, R_t) \\ &\dot{\lambda} = f_3(S, x_b^1, \lambda, x_d, R_t) \\ &R_t = f_4(S, x_b^1, \lambda, x_d) \\ &x_d^* = \frac{\int_0^{t_f} \frac{x_d^1 V}{R_t + 1} dt}{D(t_f)} = \text{purity specification} \end{aligned}$$

Discretizing the ODE's in this formulation leads to a set of nonlinear algebraic equations along with the nonlinear algebraic short-cut model to be solved simultaneously. In the resulting formulation the degrees of freedom are the design variables themselves and the control variables are determined entirely by the equality constraints. This leads to savings in computational time and memory. This formulation can also be extended to deal with overhead condenser holdup effects (equation MSM given below) by changing the modified short-cut model for simulation. Note the state variables, adjoints and the Hamiltonian remain the same and only the simulation model changes. In general, extensions can also be made to more complex models by increasing the problem size.

This formulation is valid if no inequality constraints on the control or state profiles become active, although it can easily incorporate inequality constraints on design parameters or variables evaluated at final time. Given these two approaches, we consider examples in the next section to demonstrate the use of solving the Maximum Profit Problem to simultaneously design and determine simulate the plate to plate model and the purity actually arrived at was 95.5%. The amount of distillate collected was 7.362 moles. The GAMS (Brooke *et al.*, 1988) modelling system was used for all of the models with orthogonal collocation used to discretize the differential equations.

Then the reflux policy was allowed to vary with time in order to find the optimal control policy for the Maximum Distillate Problem. The short-cut and modified short-cut models were used for the optimisation. Figure 2 compares the reflux policies and Figure 3 compares the key component purity profile.

Note that the profiles match each other closely over most of the time period. The effect of holdup is shown at the beginning and at the end of the profiles with the sharp changes in the modified holdup policies. We used the optimal control profile from the modified short-cut case (MSM) and simulated column performance with a plate to plate model. Once again we arrived at a final purity of 95.5%. Also, by allowing the reflux ratio to vary with time, we increased the amount of distillate (objective function) to 7.419 moles using the modified short-cut model. Because the no holdup short-cut model requires a higher reflux ratio for the purity specification at the beginning and end of the batch time than the modified holdup model, the amount of distillate recovered decreases to 7.259 moles. This can also be seen from equation (NT).

One sees that even though the plate to plate and short-cut models do not agree exactly because of the holdup effect, the average composition at final time agrees within 0.5%. Also for this case, the short-cut policy causes the rigorous overhead composition to be greater than the specification required and thus, the policy can be used for more complex models. We conclude that the short-cut model reflux policy can yield an overhead purity composition that will be comparable to plate to plate calculations for columns with a reasonable number of trays, nearly ideal separations, and fast column dynamics. This was also observed by Diwekar (1988).

Profit Function Maximisation

The next example we consider is found in Kerkhof and Vissers (1978) (case 13). The details of the problem are

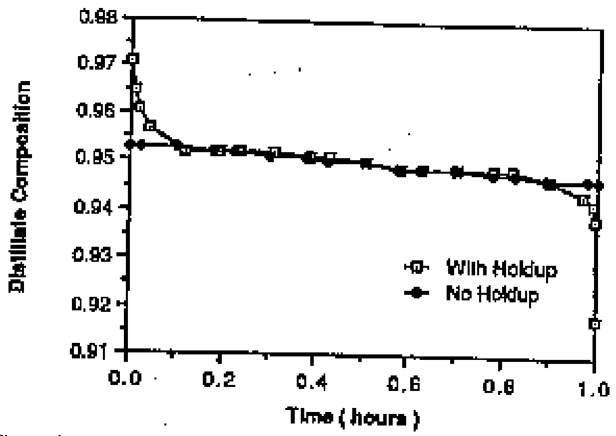

Figure 3. Distillate composition profiles for modified holdup model versus no holdup model for multicomponent example.

$N = 20$ trays, $\alpha = 2.0$, setup time T_s = one hour, overhead purity constraint = 0.98, feed composition = 0.50, ratio of sales price to cost is 41 to 10, and energy costs, depreciation, etc = 150, V = 120 moles/h, and initial charge = 100 moles. They solve the maximum profit function problem using a no holdup model, which uses quasi-stationary plate to plate calculations for the determination of the overhead composition. Here the profit function is

$$\psi = \frac{DP_r - B_0 C_0}{t_f + T_s} - C_3 \tag{30}$$

where C_3 is the cost for energy, depreciation, wages, etc.

Although our cost function considers the capital cost of the batch distillation system given above, we also solved this problem using equation (30) with the short-cut model (3) and the formulation of NLP1. The results are shown in Figure 4.

This profile compares well with the result reported by Kerkhof and Vissers. Their reflux ratio starts at about 5 and ends at slightly over 21 which compares well with Figure 4. Their optimum batch this time is slightly smaller at 3.35 h compared to our time of 3.63 h. This results in their profit being slightly higher because the product value is divided by the total batch time. Although neither model is rigorous because of the no holdup assumption, the models are useful for the preliminary design stage cost comparisons.

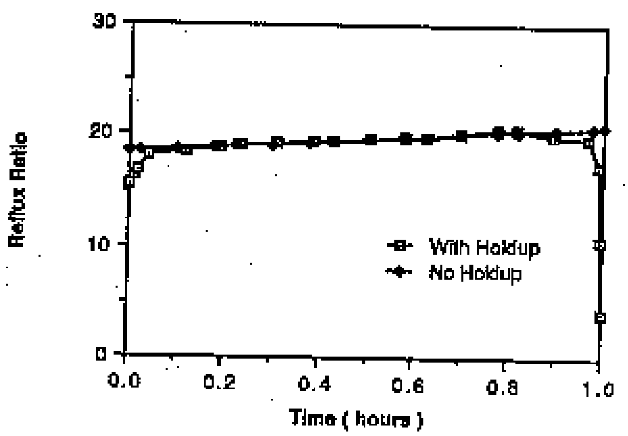

Figure 2. Comparison of reflux policies for modified holdup model versus no holdup model for multicomponent example.

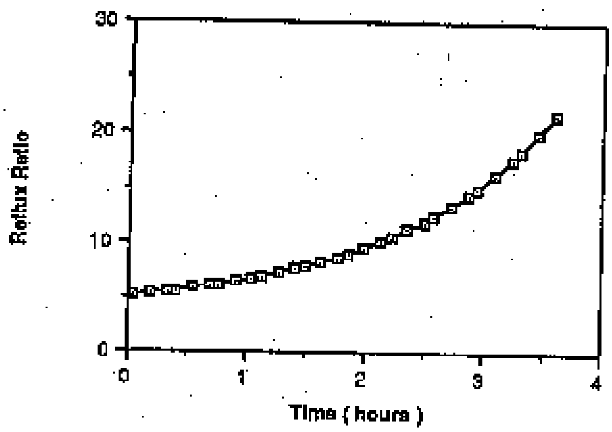

Figure 4. Optimal reflux policy for maximum profit function as defined by Kerkhof and Vissers.

separation work, i.e. trays and/or higher reflux ratio). The difference in the profit is accounted for by the smaller batch time. Again, these results are only valid for the cost parameters and objective function used in this example.

Clearly, the inequality constraint affects the final design and control profile. The collocation formulation (NLP1) allows for this constraint to be handled easily within the nonlinear program. We have shown that the design and determination of the optimal control profile can be solved for a single separation. In the next problem, we note that batch units are used for systems having different components and different feed compositions. The design of a single column and determination of the control profiles for each operation therefore needs to be considered.

Multiperiod Optimisation

Consider now the multiperiod problem for batch distillation units. This allows evaluation of a single column design for different feed compositions and services. Here we need to choose the optimal design parameters for a variety of operations and determine an optimal profile for each operation. As a small example consider the design of the column for two binary mixtures with an initial feed charge of 116 moles (each mixture) to be distilled to 95% purity. For this problem, we simplified the problem formulation by assuming instantaneous switching between the batches. Therefore, we ignored transients and downtime for the switching of the feeds. The objective function that we used was to maximise the sum of the individual profits

$$\psi_j = \frac{D_j P_{r,j} - B_{0,j} C_{0,j}}{t_{f,j}} - \frac{\text{T.A.C.}}{\text{Hrs.}}$$

for each of the periods, j. Other problem details are given in Table 5.

We initialize the solution by fixing the number of trays at 15 and solving the maximum profit function for each of the separations as shown in Figure 8.

Figure 8 clearly shows that the two mixtures require different reflux policies and the separations have different batch times. Note that the second period requires a higher reflux policy at the end because the optimal control policy tries to recover as much of the more expensive product as possible in a short amount of time. Starting from this initial design the number of trays in the multiperiod optimisation problem was allowed to vary from 5 to 20 and the maximum profit problem was resolved. For this multiperiod problem, the number of trays went to the upper bound of 20 and Figure 9 shows the solution.

Comparing this to the initial solution of 15 trays (Figures 10 and 11), we see that the reflux policy is reduced and that the batch time is also reduced. Table 6 summarizes the results.

Table 5. Input conditions—example 2.

Case	Feed composition key component	Relative Volatility	Boilup Rate moles/hr	Cost Ratio*
1	0.75	1.5	250	115/75.8
2	0.50	2.0	250	115/55.1

*Product/raw materials.

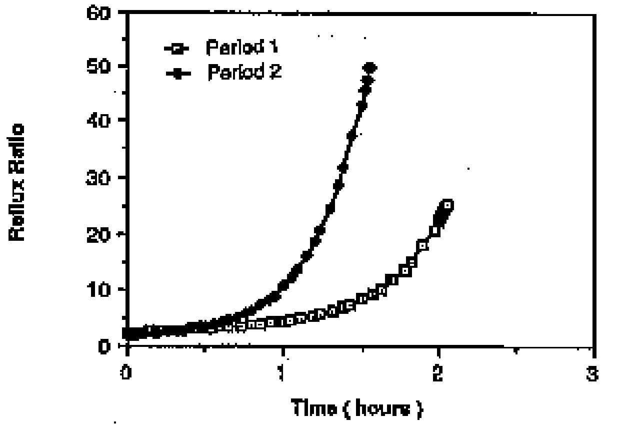

Figure 8. Optimal reflux policies for multiperiod example with $N = 15$.

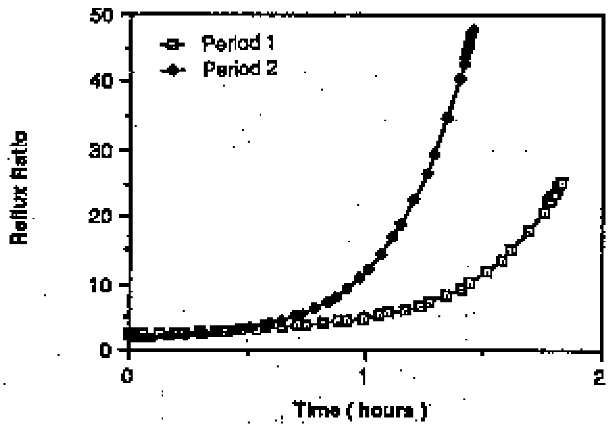

Figure 9. Optimal reflux polices for optimal design ($N = 20$).

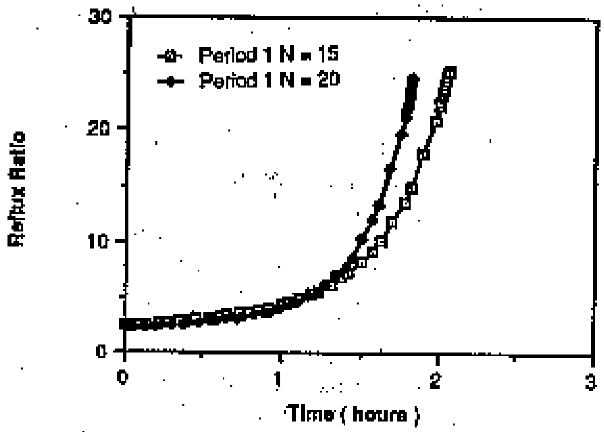

Figure 10. Comparison of optimal reflux polices for Period 1 showing the effect of number of trays on reflux policies.

For this simple case, the increase in profit is largely influenced by the decrease in the batch time. The difference in the amount of distillate recovered is slight. Note that the number of trays went to the upper bound for both of the periods. One could in principle, solve a larger number of separations by imposing a reasonable upper bound on the trays and decomposing the problem into finding the optimal reflux policy for each fixed column design. If the upper bound on the trays and boilup rate is active for each of the individual separations, then the multiperiod problem can be solved separately for each period. However, if the upper bounds are not active for

Base Case

10 tray column
V = boilup rate = 110 moles/h

From Guthrie's Correlations using Carbon Steel and Hydrocarbons as Feed Stock

Column Cost	≈ \$100,000
Reboiler, Condenser, and Kettle Cost	≈ \$200,000
Utilities	≈ \$20,000

Then

$$K_1 V^{0.5} N^{0.8} = \$100{,}000$$
$$K_1 110^{0.5} 10^{0.8} = \$100{,}000$$
$$K_1 \approx 1500$$
$$K_2 V^{0.65} = \$200{,}000$$
$$K_2 \approx 9500$$
$$K_3 V = \$20{,}000$$
$$K_3 \approx 180$$

APPENDIX B

Assume T_s is proportional to t_f, $T_s = kt_f$. (NLP1) can be written as

$$\text{(GNLP)} \quad \text{Max } \psi = \frac{DP_r - B_0 C_0}{t_f + T_s} - \frac{K_1 V^{0.5} N^{0.8} + K_2 V^{0.65} + K_3 V}{\text{Hrs.}}$$

$$\text{s.t.} \quad \dot{z} = Vf(z, y)$$
$$g(z, y) \leq 0$$
$$h(z, y) = 0$$

From (NT) we have $t_f = D/VR_{avg}$ and by defining a normalized time $\tau = Vt$ we rewrite (GNLP) equivalently as:

$$\text{Max } \psi = \frac{DP_r - B_0 C_0}{D/VR_{avg} + T_s} - \frac{K_1 V^{0.5} N^{0.8} + K_2 V^{0.65} + K_3 V}{\text{Hrs.}}$$

$$\text{s.t.} \quad \frac{dz}{d\tau} = f(z, y)$$
$$g(z, y) \leq 0$$
$$h(z, y) = 0$$
$$R_{avg}\tau_f - D = 0$$

With this formulation V appears only in the objective function. This function can now be written in a more general form:

$$\psi = \frac{a}{b/V[1+k]} - [cV^{\alpha} + dV^{\beta} + eV]$$

where $a, b, c, d, e \geq 0$ and α, β are below zero and one. Now

$$\frac{d\psi}{dV} = \frac{a}{b[1+k]} - [c\alpha V^{\alpha-1} + d\beta V^{\beta-1} + e]$$

and if we assume profitable operation, $\psi > 0$, and substitute for this inequality we have

$$\frac{d\psi}{dV} \geq [c(1-\alpha)V^{\alpha-1} + d(1-\beta)V^{\beta-1}] \geq 0$$

Thus the optimum value of V must be at its upper bound.

APPENDIX C

At a solution of (NLP1) with N fixed as a fixed parameter we can write the sensitivity of Ψ with respect to N as:

$$\frac{d\Psi}{dN} = \frac{P_r}{t_f + T_s}\frac{dD}{dN} - \frac{P_r D - C_0 B_0}{[t_f + T_s]^2}\frac{dt_f}{dN} - \frac{0.8 K_1 V^{0.5}}{N^{0.2}}$$

Here $dD/dN \geq 0$ and $dt_f/dN \leq 0$ because with an optimal reflux policy batch time can be reduced and D can be increased with increasing N. Thus a stationary point for N is only due to the third (capital cost) term.

REFERENCES

1. Barb, D. K. and Holland, C. D., 1967. *Seventh World Petroleum Congress Proceedings*, 4, 31.
2. Brooke, A., Kendrick, D. and Meeraus, A., 1988. *GAMS A User's Guide*, (The Scientific Press, Redwood City, CA).
3. Converse, A. O. and Gross, G. D., 1963. *Ind Eng Chem Fund*, 2(3), 217.
4. Cuthrell, J. E. and Biegler, L. T., 1989. *Comp & Chem Eng*, 13(1), 49.
5. Distefano, G. P., 1968. *AIChE J.*, 14(1), 190.
6. Diwekar, U. M., Malik, R. K. and Madhavan, K. P., 1987. *Comp & Chem Eng*, 11(6), 629.
7. Diwekar, U. M., 1988. *Ph.D. Thesis*, (IIT, Bombay, India).
8. Diwekar, U. M., Madhavan, K. P. and Swaney, R. E., 1989. *Ind Eng Chem Res*, 28: 1011.
9. Domenech, S. and Enjalbert, M., 1981. *Comp & Chem Eng*, 5: 181.
10. Douglas, J. M., 1988. *Conceptual Design Of Chemical Processes*, (McGraw-Hill, New York).
11. Egly, H., Ruby, V. and Seid, B., 1979. *Comp & Chem Eng*, 3: 169.
12. Guthrie, K. M., 1969. *Chem Eng*, 76(6), 114.
13. Grossmann, I. E. and Halemane, K. P., 1982. *AIChE J.*, 28: 686.
14. Huckaba, C. E. and Danley, D. E., 1960. *AIChE J.*, 6: 335.
15. Kerkhof, L. H. J. and Vissers, H. J. M., 1978. *Chem Eng Sci*, 33: 961.
16. Logsdon, J. S. and Biegler, L. T., 1989. *Ind & Eng Chem Res*, 28(11), 1628.
17. Luyben, W. L., 1971. *Ind Eng Chem Process Des Develop*, 10(1), 54.
18. Mayur, D. N. and Jackson, R., 1971. *Chem Eng J*, 2: 150.
19. Meadows, E. L., 1965. *Chem Eng Prog*, (sym. ser.), 46: 48.
20. Mujtaba, I. M. and Macchietto, S., 1988. *Proc 12th IMACS World Congress, Paris, July 18–22.*
21. Mujtaba, I. M. and Macchietto, S., 1988. *SIMO'88-Simulation et optimisation en Genie des Procedes, Toulouse (France), Sept. 14–15.*
22. Pigford, R. L., Tepe, J. B. and Garrahan, C. J., 1951. *Ind Eng Chem*, 43: 2592.
23. Robinson, E. R., 1970. *Chem Eng Sci*, 25: 921.
24. Vasantharajan, S. and Biegler, L. T., 1988. *Comp & Chem Eng*, 12(11), 1089.
25. Villadsen, J. and Michelsen, M. L., 1978. *Solution of Differential Equation Models by Polynomial Approximation* (Prentice-Hall, Englewood Cliffs, NJ).
26. Viswanathan, J. and Grossmann, I. E., *Comp & Chem Eng* (to appear).

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