

1992

Accurate determination of optimal reflux policies for the maximum distillate problem in batch distillation

Jeffery Scott. Logsdon
Carnegie Mellon University

Lorenz T. Biegler

Carnegie Mellon University. Engineering Design Research Center.

Follow this and additional works at: <http://repository.cmu.edu/cheme>

This Technical Report is brought to you for free and open access by the Carnegie Institute of Technology at Research Showcase @ CMU. It has been accepted for inclusion in Department of Chemical Engineering by an authorized administrator of Research Showcase @ CMU. For more information, please contact research-showcase@andrew.cmu.edu.

NOTICE WARNING CONCERNING COPYRIGHT RESTRICTIONS:

The copyright law of the United States (title 17, U.S. Code) governs the making of photocopies or other reproductions of copyrighted material. Any copying of this document without permission of its author may be prohibited by law.

**Accurate Determination of Optimal Reflux Policies
for the Maximum Distillate Problem in Batch Distillation**

J.S. Logsdon, L.T. Biegler

EDRC 06-134-92

Accurate Determination of Optimal Reflux Policies for the Maximum Distillate Problem in Batch Distillation

J. S. Logsdon and L. T. Biegler**
Department of Chemical Engineering
Carnegie Mellon University
Pittsburgh, PA 15213

An approach for the accurate solution of optimal control problems that arise in batch distillation is developed and demonstrated. Since the optimal control problem has a natural partitioning of control variables and state variables, we develop a nonlinear programming decomposition strategy to (1) exploit the block matrix form of the discretized differential equations that results from using collocation on finite elements, and (2) perform the optimization in the reduced space of the control variables. State variables for each finite element are determined by linearized differential equations and information is passed from element to element by chainruling the state information. In addition, the nonlinear programming strategy has a great deal of flexibility to determine control variable discontinuities and enforce a wide variety of state and control variable constraints.

In this study, we also consider characteristics of the maximum batch distillate problem and show that our approach is especially useful for the optimization of detailed tray-by-tray models with tray and condenser holdups. Here we discuss two formulations: an inequality path constrained problem and the classical endpoint constrained problem. In both cases interesting and unusual optimal policies are determined and compared to current practice. Moreover, parallels are observed between optimal reflux policies for these two problems, and these are also related to findings from previous studies. To handle these problems, nonlinear programs of up to 8000 variables are solved reasonably quickly on a small workstation. Finally, it is observed that more complex batch distillation problems can be handled in a straightforward manner through this approach.

Submitted to I & EC Research
April, 1992

* to whom all correspondence should be addressed

+ JL S. Logsdon is currently with Amoco Chemical Corp., Naperville, IL

1. Introduction

The determination of optimal control profiles for large chemical processes described by differential and algebraic equations (DAEs) remains a challenging problem. In particular, DAE optimization methods must handle state and control variable (equality and inequality) constraints. In the simultaneous approach described here, the DAE system is converted to a set of algebraic equations using orthogonal collocation on finite elements, and one can deal with state path constraints and control path constraints simply by including them in the nonlinear programming formulation. This approach has been used by Cuthrell and Biegler (1987,1989), Renfro et al. (1987), Eaton et al. (1988), and Logsdon and Biegler (1989) to address the optimal control problem.

While the simultaneous approach offers a number of advantages for dynamic optimization problems, the resulting nonlinear programming formulations for these problems can become large. Consequently, problem structure must be exploited in order to solve the resulting NLP problem efficiently. Vasantharajan and Biegler (1988) and Vasantharajan et al. (1990) develop a general purpose decomposition algorithm for Successive Quadratic Programming (SQP) and demonstrate its efficiency and reliability with respect to other general purpose NLP solvers. Logsdon et al. (1990) also applied this approach to the optimal operation of batch distillation systems. More recently, Logsdon and Biegler (1992) develop several algorithms tailored to the block lower-triangular structure of the collocation equations.

In this study, we apply this structured decomposition algorithm to the maximum distillate problem for batch columns and also relate this problem to more general optimization problems for batch processing. Here we consider tray-by-tray distillation models with tray and condenser holdups. With an accurate optimal reflux policy for more realistic models, we also compare these results to current operating policies and methods of analysis. In the next section we provide a brief review of previous work. Then, in section 3 we outline the general NLP formulation for optimal control problems and briefly sketch a structured, reduced gradient optimization approach tailored to this problem class. This approach is based on concepts of finite element collocation and Successive Quadratic Programming (SQP). Section 4 discusses the maximum distillate problem and presents some interesting optimal reflux policies for path constrained and endpoint constrained maximum distillate problems. The solutions of these two process examples are also compared to constant

optimal reflux policies and piecewise constant reflux policies; significant differences and advantages are observed with our approach. Also, we briefly discuss extensions of the maximum distillate problem to more complex problem classes. Finally, in Section 5 we draw conclusions and discuss directions for future work.

2. Batch Distillation Literature Review

The determination of distillation policies for batch columns has received considerable attention since the 1940's. Early work concentrated on the development of approximate methods for dynamic simulation of these columns (see the review by Luyben (1971)). Batch distillation models are composed of differential and algebraic equations which are the mass and energy balances, the thermodynamic relationships, and the dynamic plate relationships. Short cut methods have also been used to reduce the size of the systems of the DAE's; otherwise the number of equations increases with the number of components and number of plates under consideration.

Optimization studies have been reported for binary systems by Luyben (1971), Coward (1967), Robinson (1971), Mayur and Jackson (1971), and Keikhoff and Vissers (1978), and for ternary systems by Luyben (1988). The optimal control variable is usually the reflux ratio, since it can be shown that the optimal boilup rate frequently remains at its upper bound (see discussion in section 5). Control profiles have been reported for various optimization studies classified according to the objective functions being considered. Normally problems found in the literature are defined as follows:

1. **Maximum Distillate Problem** - Maximize the amount of distillate of a specified concentration for a specified batch time.
2. **Minimum Time Problem** - Minimize the batch time needed to produce a prescribed amount of distillate of a specified concentration.
3. **Maximum Profit Problem** - Maximize a profit function for a specified concentration of distillate.

Converse and Gross (1963) first solved the maximum distillate problem using dynamic programming and Pontryagin's maximum principle. The minimum time problem has been studied by Coward (1967), Robinson (1970) and Mayur and Jackson (1971) and the

maximum profit problem has been studied by Kerkhof and Vissers (1978). Murty et al (1980) compare several optimal control methods for batch distillation optimization while Hansen and Jorgensen (1986) apply orthogonal collocation to solve the (unconstrained) optimality conditions. Most of the work found in the literature is restricted to binary mixtures with the assumptions of no holdup on the trays, and fixing column design variables such as the number of trays and vapor boilup.

More recently, Diwekar et al. (1987), Mujtaba and Macchietto (1988), Farhat et al. (1990) and Logsdon et al. (1990) have investigated batch distillation as optimization problems for multicomponent systems. Diwekar et al (1987) used shortcut models along with Pontryagin's maximum principle to obtain control policies for the maximum profit problem. Mujtaba and Macchietto (1988) considered tray to tray dynamics and included material and energy balances, plate holdup, and rigorous phase equilibrium to solve the maximum distillate problem. They also considered more general problems that involve the recycling of intermediate distillation cuts, as did Faihat et al. (1990). Logsdon et al (1990) used the shortcut model to simultaneously optimize the design of the column and to obtain the optimal reflux policy to maximize a profit function, which includes capital costs along with operating costs.

For the design of batch distillation systems, Al-Tuwaim and Luyben (1991) developed a shortcut design method to determine the number of trays and reflux ratio. Implicit in this approach is a constant reflux policy (see also Luyben (1971, 1988)). Diwekar and Madhavan (1991), on the other hand, develop the BATCH-DIST package which incorporates various levels of complexity for batch distillation models, from Fenske-Underwood-Gilliland correlations to tray-to tray batch columns models. In many cases, the shortcut models compare quite well with more rigorous simulations and lead to savings of up to two orders of magnitude in computational effort. This approach also allows for different optimization problem formulations as well. Finally, Mujtaba and Macchietto (1991) studied the effects of tray and condenser holdups on batch distillation optimization. Assuming constant reflux ratios they obtained optimal holdups and determined conditions under which holdups can improve column performance.

In this work we consider a tray-by tray holdup model for batch distillation and study the influence of the optimal reflux policy. As mentioned above, our approach determines accurate optimal reflux policies under a variety of constraints and these are exploited to uncover some unusual behavior. These are then compared to optimal policies determined

by other approaches in order to demonstrate the effectiveness of our procedure. Also, we investigate the characteristics of the optimal solution of higher "index" systems which are often bypassed with simpler tray dynamics and reflux policies.

3. NLP Formulation

In this section we briefly review the NLP formulation with collocation on finite elements. Consider the following general optimization problem over $t \in [a,b]$:

$$\begin{aligned}
 & \text{Min}_{\mathbf{u}(t), \mathbf{z}(t), \mathbf{p}} \quad \int_a^b G(\mathbf{z}(t), \mathbf{u}(t), \mathbf{p}) \, dt & (1) \\
 & \text{s.t.} \quad \dot{\mathbf{z}}(t) = \mathbf{F}(\mathbf{z}(t), \mathbf{u}(t), \mathbf{p}) \\
 & \quad \mathbf{g}(\mathbf{u}(t)) \leq \mathbf{0} \\
 & \quad \mathbf{g}_f(\mathbf{z}(b)) \leq \mathbf{0} \\
 & \quad \mathbf{z}(a) = \mathbf{z}_0 \\
 & \quad \mathbf{z}(t)^L \leq \mathbf{z}(t) \leq \mathbf{z}(t)^U \\
 & \quad \mathbf{u}(t)^L \leq \mathbf{u}(t) \leq \mathbf{u}(t)^U
 \end{aligned}$$

where:

$\int_a^b G(\mathbf{z}(t), \mathbf{u}(t), \mathbf{p}) \, dt$ = objective function terms evaluated at final conditions

$G(\mathbf{z}(t), \mathbf{u}(t), \mathbf{p})$ = time dependent objective function term

$\mathbf{z}(t)$ = state profile vector

$\mathbf{u}(t)$ = control profiles

\mathbf{p} = design parameters, not time dependent

\mathbf{g} = inequality design constraint vector

\mathbf{g}_f = inequality constraints at final conditions

\mathbf{z}_0 = initial condition for state vector

$\mathbf{z}(t)^L, \mathbf{z}(t)^U$ = state profile bounds

$\mathbf{u}(t)^L, \mathbf{u}(t)^U$ = control profile bounds

Here, we discretize the DAE system by using a piecewise polynomial approximation on finite elements and by applying orthogonal collocation to construct the algebraic residual equations. These residuals are evaluated at the shifted roots of an orthogonal Legendre polynomial. Consider now the initial value problem over a finite element, i , with time

¹⁸ [$C_i > C_{i+1}$] with state and control profiles approximated by Lagrange-type polynomials over this element. Recall that the Lagrange polynomial has the desirable property that, for $z_{K+1}(t_{ij}) = z_{ij}$ (0, for example,

$$z_{K+1}(t_{ij}) = z_{ij}, \quad (2)$$

This polynomial also allows for the direct imposition of path constraints within the problem formulation. Applying K point, orthogonal collocation on finite elements and defining the basis functions so that time is normalized over each element, one obtains the following residual equation for the ODE:

$$\Delta \zeta_i r(t_{ik}) = \sum_{j=0}^K z_{ij} \dot{\phi}_j(\tau_k) - \Delta \zeta_i F(z_{ik}, U_{ik}) = 0 \quad (3)$$

$$i = 1, \dots, NE$$

$$k = 1, \dots, \bar{K}$$

where the basis function $\phi_j(\tau_k) = \frac{d\phi_j}{d\tau}$ is calculated offline and $\tau = \xi + A\Delta t_k$. This form is convenient to work with when the element lengths are included as decision variables, because it is still defined even if $A\Delta t$ goes to zero during the solution of the optimization problem. Note also that element lengths play two roles. First, they need to be sufficiently small in order to allow the piecewise polynomials to approximate the solution profiles accurately. Second, element lengths locate points of discontinuity for the control profiles. In our NLP formulation, we enforce the continuity of the states at element boundaries, but allow the control profiles to have discontinuities at these endpoints. These endpoints also provide the initial conditions for the next element states.

Discretization of the optimal control problem leads to the NLP problem given below. This formulation consists of the discretized DAE model, the continuity equations for state variables, and any additional equality and inequality constraints in the problem formulation. Finally, approximation error constraints are added that ensure the accuracy of the solution profiles. These can range from simple bounds on element lengths to constraints based on detailed approximation error estimates.

$$\begin{aligned}
 & \text{Min} \quad \Psi(z_f, p) + \sum_{i=1}^{NE} \sum_{j=1}^K w_{ij} G(z_{ij}, u_{ij}, p, \Delta\zeta_i) \\
 & \text{st} \quad \text{AC}_i r_{ij} = iK + i r_{t_{ij}} - A \&F(Z_{ij}, U_{ij}, p) = 0 \\
 & \quad g(z_{ij}, u_{ij}, \Delta\zeta_i) \leq 0 \\
 & \quad gf(Z_f) \wedge 0 \\
 & \quad z_{i0} - z_0 = 0 \\
 & \quad z_{i0} - z_{i1} (CO = 0 \quad i = 2, \dots, NE) \\
 & \quad z_f - z_{K+1}^{NE} (\zeta_{NE+1}) = 0 \\
 & \quad L \leq z_{ij} \leq U \\
 & \quad L \leq u_{ij} \leq U \\
 & \quad A \wedge AC_j \leq \Delta U \\
 & \quad \sum_{i=1}^{NE} \Delta\zeta_i = \zeta_{Total}
 \end{aligned} \tag{4}$$

Variables in (4) include AC_i finite element lengths for $i=1, \dots, NE$; z_f , the value of the state at the final time; z_{ij} and u_{ij} , the collocation coefficients for the state and control profiles, respectively; and p , any additional design parameters (such as boilup rate and final time). Finally, the order of the collocation method is often determined by the state variable constraints. Activity of these constraints over a nonzero time period often increases the index of the DAE system; the collocation method must therefore have stability and accuracy properties to handle these cases (Logsdon and Biegler, 1989).

To solve (4), we note that for given values of control variables and element lengths one can determine the state variable trajectories from the collocation equations and pass the information from element to element. Once the trajectories have been computed, and derivative information (sensitivity of states to control variables) is obtained, we chainrule this information in order to obtain the *reduced gradients* of the objective and constraint functions. We then update the optimal control profile through a reduced space Successive Quadratic Programming (SQP) algorithm: Here the collocation equations are converged for each SQP iteration and the resulting method is a feasible path, reduced gradient approach. Note however, that the constraints that govern the approximation error and the element

lengths are still manipulated at the optimization level. Further details of this approach and its infeasible path variations can be found in Logsdon and Biegler (1992). A brief description of the algorithm is presented next.

Reduced Gradient Algorithm:

0. Choose the number of elements and the corresponding number of collocation points based on the likely index of the DAE system (see Logsdon and Biegler, 1989, for details). Initialize the control variables, state variables, and element lengths.
1. For values of the control variables and element lengths at iteration k , and initial conditions for the state variables, perform the following for each element i ($i = 1, \dots, NE$):

- 1.1 Using the initial conditions of element i as starting guesses, solve the residual equations (3) by a Newton algorithm to obtain the interior states z_i^k . Here A represents the Jacobian of (3) with respect to z_i^k and b represents the right hand sides of (3).

- 1.2 Calculate the derivatives for this element's decision variables:

$$\frac{\partial z_i^k}{\partial t} = A^{-1} \left[\frac{db}{dt} \frac{dA}{dz_i^k} \right]$$

where t represents the control variables and the element length for each

finite element. Note that $\left[\frac{\partial b}{\partial t} \frac{dA}{dz_i^k} \right]$ can be determined analytically from the differential equations. A^{-1} is available from the Newton step in 1.1.

- 1.3 Apply the state continuity equations, and solve for the next element's initial conditions:

$$z_i^k = z_{i+1,0} = \sum_{j=0}^K z_{ij} \phi_j(\tau=1) \quad (5)$$

- 1.4 Chainrule the derivatives from previous elements and update:

$$\frac{dz_i^*}{d\xi_j} = \frac{dz_i^*}{dz_{i-1}^*} \frac{dz_{i-1}^*}{dz_{i-2}^*} \dots \frac{dz_i^*}{d\xi_j} \quad (6)$$

2. Continue until an intermediate element is reached that influences an inequality ($g(z_c)$) or until the last element is reached. Determine the reduced gradients for the objective and constraint functions according to equation (7).

$$\{Z^T \nabla \Phi\}_j = \frac{\partial z_i^*}{\partial \xi_j} \frac{\partial \Phi}{\partial z_i} \quad \{Z^T \nabla g_n\}_j = \frac{\partial z_c^*}{\partial \xi_j} \frac{\partial g_n}{\partial z_c}, \quad n = 1, n_g \quad CD$$

3. Assemble the objective and constraint function values and reduced gradients from the above steps.
4. Call the SQP algorithm (Cuthrell and Biegler, 1985). If Kuhn-Tucker conditions are satisfied, Stop. Otherwise the algorithm solves the following quadratic program at iteration k :

$$\begin{aligned} \text{Min}_{x^*} \quad & \text{VO}^T Z A \xi + \frac{1}{2} \xi^T (Z^T B Z) A \xi \\ \text{s.t.} \quad & g + \nabla g^T Z A \xi \leq 0 \end{aligned} \quad (8)$$

to determine the search direction, $A\xi$. Note that this QP contains all of the state and control variable inequality constraints. In addition, SQP also updates the reduced Hessian matrix, $(Z^T B Z)$, based on the BFGS formula, and performs a line search to determine the steplength for the decision variables, u and $A\xi$ (see Cuthrell and Biegler, 1985).

5. Return to step 1, with a new set of decision variables from SQP.

This feasible path algorithm can be viewed as a hybrid between the nested approach (as used by Mujtaba and Macchietto, 1988) and the simultaneous approaches. Like the nested approach, a small subproblem is considered at the NLP level and the equality constraints from the DAE discretization are solved in an inner loop (step 1.1). The algorithm also inherits all of the convergence properties of the SQP method in the decision variable space. On the other hand, this algorithm also controls the element lengths at the NLP level and

allows for the straightforward application of state and control variable constraints, as with the simultaneous method.

4. Batch Distillation Examples

Chemical engineering processes described by staged models of differential and algebraic equations frequently form large optimization problems. To demonstrate the performance of our algorithm, we investigate a simple toluene/cyclohexane system which has been successfully simulated by Domenech and Enjalbert (1974) and by Cuille and Reklaitis (1986). We consider the maximum distillate problem for a batch time of one hour. The simulation model described by Domenech and Enjalbert (1980) for the column model, which assumes equal heats of vaporization and heat capacities for the components, is used for this problem. More rigorous column models are also described by Cuille and Reklaitis (1986) and Mujtaba and Macchietto (1988). Details of the example are as follows:

V, boilup rate = 120 moles/hr
 N= 10 trays
 H_p, plate holdup = 1 mole
 H_{con}* condenser holdup = 1 mole

Initial Concentrations:

X^Q = 0.55, cyclohexane
 x_{b,0}² = 0.45, toluene

The optimal control problem can be written as:

$$\text{Maximize} \quad V = \text{Dis} = D(t_f) \quad (9)$$

$$Rt(t)$$

s.t.

Mass Balances:

$$\frac{dS}{dt} = -\frac{v}{R_j + 1}, \quad S = \text{overall mass} = S_0 - D(t)$$

$$\frac{dx_b^j}{dt} = \frac{V}{S} \left[r_b^j - y_b^j \frac{[R+1]}{[R+1]} (x_b^j - x_b^j) \right] \quad *i = \text{component} \text{ in pot}$$

$$\frac{dx_p^j}{dt} = \frac{V}{S} \left[P_i^j y_i^j - [R+1] x_p^j \right] \quad 4 = \text{component } j \text{ on plate } p$$

$$\frac{dx^j}{dt} = -\frac{V}{H_{con}} [y^j - x_{j+1}^j], \quad j = \text{component } j \text{ in condenser accumulator}$$

Vapor-Liquid Relations (Here K values are modeled by ideal vapor pressure relations using Antoine equations and data from Reid et al, 1977.)

$$\sum_{j=1}^{NC} y_p^j = 1.0$$

$$y_p^j = K_p^j x_p^j$$

$$K_p^j = f(T, x_p^j)$$

Purity constraints for first component:

$$x_{d, \text{lower bound}}^j = \frac{\int_{t_0}^{t_1} y_{d, \text{lower bound}}^j dt}{\int_{t_0}^{t_1} \frac{V}{RHT} dt} \geq 1.0 \quad \text{time average purity constraint}$$

or

$$x_p^j, \text{ lower bound} \leq x_p^j \leq 1.0 \quad \text{constant purity constraint on plate } p$$

Note that if the constant overhead problem is solved with the purity specification enforced on the distillate ($x_{d, \text{lower bound}}^j \geq x_d^j$) an index 3 DAE system can result. Here the reflux ratio, R_t , can be taken as the index variable. The problem becomes index 3 whenever the overhead purity constraint is active, and either a high order, (strongly A-) stable method must be applied or the constraint is reformulated to one of lower index. To see that this system is indeed index 3, we differentiate the purity constraint with time as follows:

$$x_{d, \text{lower bound}}^j = x_d^j \quad \text{Index 3}$$

$$\frac{dx_d^j}{dt} \text{ lower bound} = \frac{dx_d^j}{dT} = \frac{V}{H_{con}} \quad , \quad j \quad j \quad , \quad \text{Index!}$$

$$\begin{aligned} \frac{d^2 x_d^j}{dt^2} &= \frac{d^2 x_{N+1}^j}{dt^2} = \frac{V}{H_{con}} \left[\frac{dy_N^j}{dt} - \right] \quad \text{at} \quad \text{Index 1} \\ &= \frac{V}{H_{con}} \left[K_p^j \frac{dx_N^j}{dt} - \frac{V}{H_{con}} [y_N^j - x_{N+1}^j] \right] \end{aligned}$$

Note that since the reflux ratio is explicit in the expression for $\frac{dx^j}{dt}$, we require one more differentiation to obtain an index 0 expression.

$$\begin{aligned} \frac{dx_N^j}{dt} &= \frac{V}{H_p} \left[y_{N-1}^j - y_N^j + \frac{R_t}{[R_t + 1]} (x_{N+1}^j - x_N^j) \right] \\ \frac{d^2 x_N^j}{dt^2} &= f \left[\frac{dR_t}{dt}, \frac{dy_{N-1}^j}{dt}, \frac{dy_N^j}{dt}, \frac{dx_{N+1}^j}{dt}, \frac{dx_N^j}{dt} \right] \quad \text{Index 0} \end{aligned}$$

To avoid numerical difficulty, we can reduce the constant overhead problem to index 2 by enforcing a corresponding purity constraint on the top plate instead of on the distillate composition, as seen below:

$$\begin{aligned} x_N^j \text{ lower bound} &= x_N^j \quad \text{Index 2} \\ \frac{dx_N^j}{dt} &= \frac{V}{H_p} \left[y_{N-1}^j - y_N^j + \frac{R_t}{[R_t + 1]} (x_{N+1}^j - x_N^j) \right] \quad \text{index 1} \\ \frac{d^2 x_N^j}{dt^2} &= f \left[\frac{dR_t}{dt}, \frac{dy_{N-1}^j}{dt}, \frac{dy_N^j}{dt}, \frac{dx_{N+1}^j}{dt}, \frac{dx_N^j}{dt} \right] \quad \text{Index 0} \end{aligned}$$

On the other hand, for the time average purity (endpoint) constraint, the system is only index 1 and no reformulation is required. For this example, we investigate both the path (index 2) and the endpoint purity constraint problems. We specify the top plate purity constraint as:

$$x_N^1, \text{ lower bound} = 0.995 \leq x_N^1$$

which corresponds to a distillate path constraint purity of 0.998. For the endpoint constraint, we specify the average distillate purity as:

$$x^d, \text{ lower bound} = 0.998 \leq \bar{x}_d = \frac{\int_0^{t_r} R_{t+i} dt}{\int_0^{t_r} \frac{V}{R_t+1} dt} \leq 1.0$$

In addition, we have 2 components and 12 stages (10 trays with reboiler and condenser) leading to a system of 24 differential material balance equations plus an overall material balance. Here we set up the total reflux system of equations to solve for the initial conditions, which are constants in our optimization problem. The total reflux initialization is used for this example as it is typical for a high purity separation. In other applications, e.g., where equilibration time for total reflux is significant, other initial conditions, such as cold starts or partial equilibration, may be more appropriate. Our approach handles these equally well.

Using three point collocation for the discretization, we define the following state and control variables at each of the interior collocation points as follows:

	$R_{t,1}$	$R_{t,2}$	$R_{t,3}$	
	$y_{p,1}^j$	$y_{p,2}^j$	$y_{p,3}^j$	
	$K_{p,1}^j$	$K_{p,2}^j$	$K_{p,3}^j$	
	$T_{p,1}$	$T_{p,2}$	$T_{p,3}$	
$x_{p,0}^j$	$x_{p,1}^j$	$x_{p,2}^j$	$x_{p,3}^j$	$x_{p,n}^j$
S_0	S_1	S_2	S_3	S_{n+1}
I	*	*	*	1
C_0				C_1

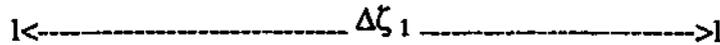


Figure 1: Finite element 3 point collocation for batch distillation

- y_j , = vapour compositions
- x_p^j = liquid compositions
- K_j , = equilibrium constant
- T_p = temperature on plate
- S = overall mass = $S_0 - D(t)$
- R_t = reflux ratio

We write explicit expressions for the derivative terms and the right hand sides of the differential equations at each of the collocation points. This requires expressions for the equilibrium constants, and additionally bubble point constraints must be enforced. To complete the model, we add the continuity equations for x_p^j and for S . The resulting NLP

has 429 constraints and up to 457 variables for each element

4.1 Path Constrained Problems

We first consider the path constraint purity for the top plate. In order to obtain a starting profile for this problem, we solve the maximum distillate problem (for the constant overhead constraint) sequentially for each element by specifying the initial conditions and the integration length and marching forward (see Figure 2).

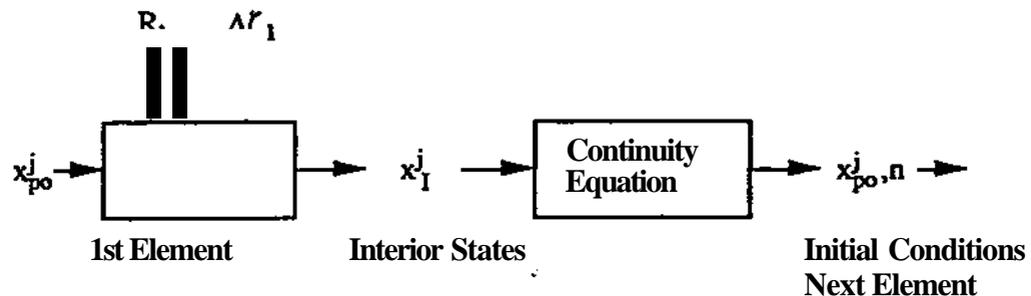


Figure 2: Solution of collocation equations within one finite element

This is similar to the element-to-element approach described in the above algorithm, but now we are actually optimizing the control profile within each element. This initialization

strategy allows the modeler to estimate the number of finite elements needed for accurate integrations; regions that require small integration lengths can be identified for the initialization of the larger NLP. Additionally, it is easier to verify and debug this smaller NLP before the finite elements are linked **together**.

Solving the maximum distillate problem for each of 18 elements requires a total of 7722 constraints and 8208 variables. Following this initialization and starting profile, we now set up the column model with the decomposition algorithm in section 3. Here, we obtain an accurate optimal reflux policy with only 12 finite elements as shown in Figure 3. A performance comparison of this approach is given in Table L

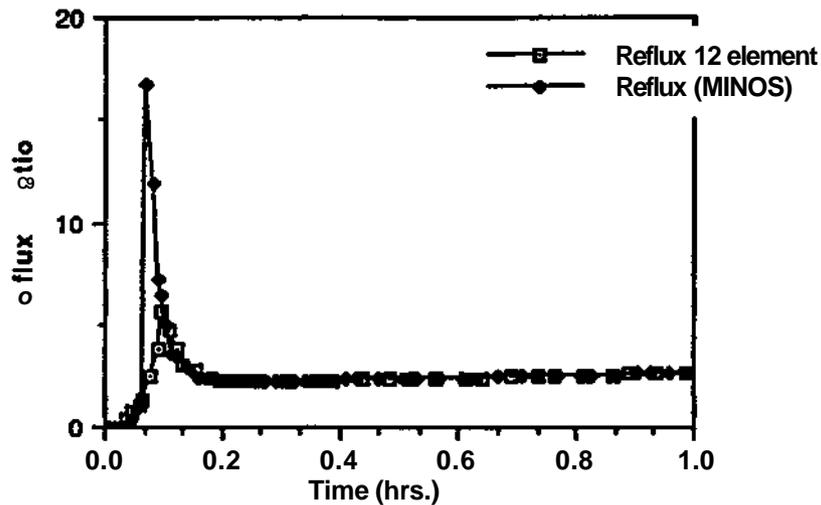


Figure 3: Comparison of reflux policies: MINOS element-by-element initialization and optimal policy determined by SQP (with 12 elements).

Table 1: Comparison of Element to Element Optimization (MINOS) with Entire Batch Time Optimization (SQP) - TopPlate Purity Constraint Problem

Total Distillate Recovered (moles)	Number Finite Elements	Total CPU time (hrs: mins: sees)
(MINOS)	1	

38.591	(optimized 18 times)	0:20:19
(SQP)		
38.615	12	1:09:44

Note that the amount of distillate recovered is dominated by the active path constraint. This allows the control profile from the element-to-element optimization approach to be close to the optimal control profile. It is also interesting that the initial policy of total takeoff (or zero reflux policy) was also identified by Coward (1967)- In Coward's examples, this policy was demonstrated numerically to be superior for easy separations where the initial distillate has a higher purity than the purity specification. While Coward used a no holdup model for his work and commented that the "zero reflux" policy should not be needed for most separations, his zero reflux observation coincides precisely with the behavior shown in Figure 3. Also, to initialize their constant overhead policies, Diwekar and Madhavan (1991) suggest a zero reflux policy as well.

Moreover, because we have included holdup for the column model, this effect is more pronounced because the amount of high purity material to draw upon is present longer in the top section of the column because of the "fly wheel" effect (Luyben, 1971; Pigford et al., 1951). This can be seen from Figure 4. Here the reflux policy maximizes the distillate takeoff as long as the top plate purity exceeds the purity constraint (0.995). Then the reflux policy quickly increases to meet the purity of the top section of the column. Once the path constraint becomes active, it determines the reflux policy for the remainder of the batch time.

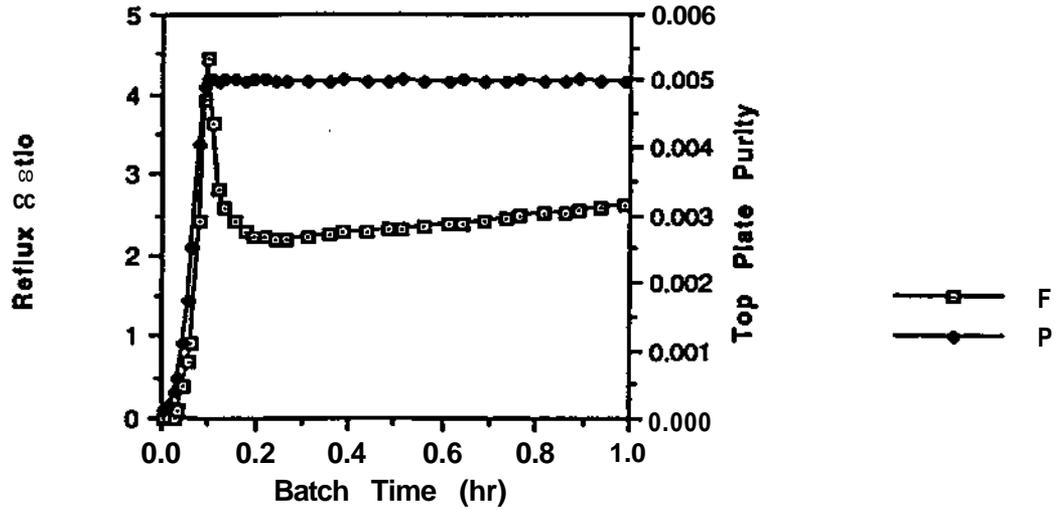


Figure 4: Optimal Path Constrained Policy and Purity ($1 - x_j$) Profiles, Note that a Top Plate Purity of 0.995 Corresponds to Distillate Purity of 0.998. The open squares indicate the reflux policy (left axis), the closed diamonds indicate the purity profile.

4.2 Endpoint Purity Constrained Control Problem

The previous case shows how our algorithm can be used to construct and efficiently determine optimal control profiles for problems that have active path constraints and require a large number of state variables. We now consider the classic optimal control problem which is governed by the time average (endpoint) purity constraint

$$x_{d, \text{lower bound}} = 0.998 \leq \bar{x}_j = \frac{\int_0^{t_f} \frac{x_j^* V}{R_t + 1} dt}{\int_0^{t_f} \frac{V}{R_t + 1} dt} < 1.0$$

The problem definition and initial conditions are the same as for the constant overhead case presented earlier. In order to determine a good initialization for the NLP, we solve the optimal control problem using the *shortcut model* developed by Diwekar (1988). Here the

relative volatility ($\alpha = 2.48$) was found from the geometric average of the reboiler relative volatility and the top plate relative volatility at total reflux as described by Diwekar (1988). The shortcut model is a much smaller system of differential equations (3 differential equations) than our stage-to-stage model (24 differential equations). The optimal policy is shown in Figure 5 for four finite elements with fixed integration lengths of 0.25 hrs. This reflux ratio policy produces 37.029 moles of distillate. The requirements for MINOS to solve this NLP (223 constraints, 255 variables) are 132.8 sees, for 34 iterations on a Vax 3200.

The shortcut model requires only four elements to find an optimal control profile because it considers only the differential equations for the overall material balance and the reboiler composition differential equations (see Logsdon et al, 1990). However, if we wish to consider tray dynamics accurately for the stage-to-stage column model, we require smaller integration steps and more finite elements. From Logsdon et al. (1990) we know that the profile will be active at the beginning and end of the batch time because of the holdup effect; smaller finite elements are required here because the control profile will be steeper at these times. Additionally, we can estimate the size of the finite elements from the constant overhead case, which required 12 elements. Here we set the number of elements to 16 and start with a flat control profile of 3.0 for the reflux ratio. Since we are not directly enforcing the residual equations for each of the stages we allow the integration lengths to vary slightly in order to obtain accurate approximations.

Additionally, we enforce accuracy constraints on the the control profile by enforcing its lower and upper bounds at the breakpoint elements using simple extrapolations. Also, a small tolerance on the K th derivative of the control profile is enforced (see Logsdon and Biegler, 1989) within each element. The QP for 16 finite elements includes an equality constraint (the sum of the integration lengths equals 1 hour) and 81 inequality constraints for the accuracy constraints and the endpoint purity constraint. The number of variables is 64 ($NE*4$) variables, 455 within each element and a total of 7280 variables. Using our algorithm we arrive at the control profile shown in Figure 5. Figure 6 shows the purity constraint along with the control profile.

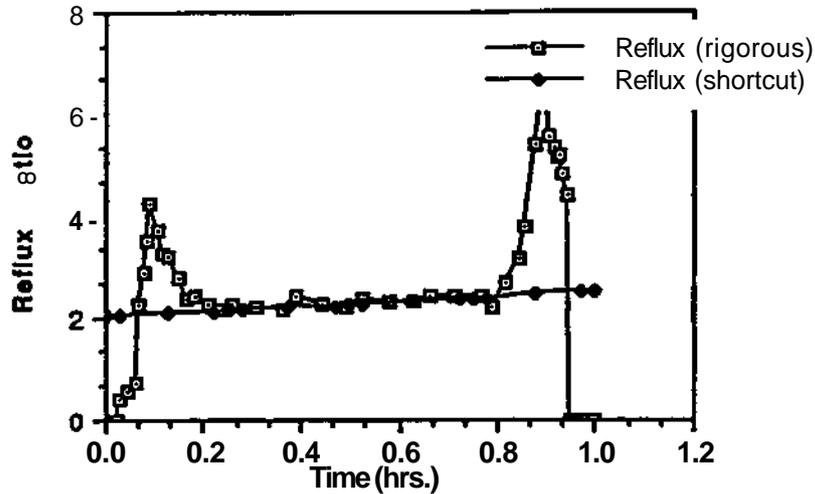


Figure 5: Comparison of shortcut model control profile versus stage-to-stage model for the endpoint constrained problem.

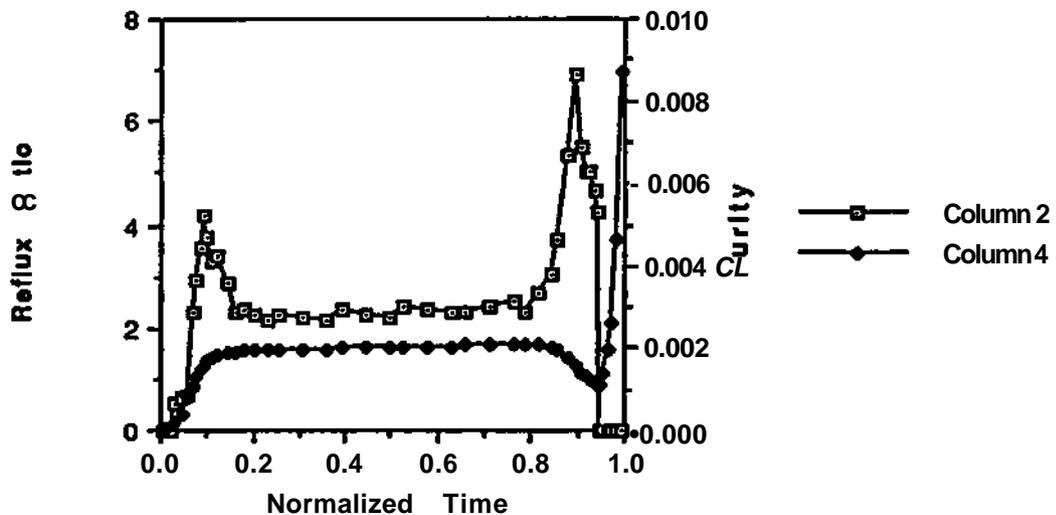


Figure 6: Optimal Endpoint Constrained Policy and Purity ($1 - x_j$) Profiles. The open squares indicate the reflux policy (left axis), the closed diamonds indicate the purity profile..

In this figure we clearly see the effect of holdup on the optimal reflux policy. Again, the policy is to run the column at maximum distillate takeoff rate at the beginning and end of the batch time. Then in order to maintain the overall distillate purity, the distillate composition must be concentrated near the periods of maximum takeoff, resulting in the profile shown. Initially, this is similar to the policy for the path constraint problem.

However, with the accumulated distillate and the aim of "dumping" the holdup at the end, the column runs at zero reflux at the end of the distillation period.

It should be noted that we solved the maximum distillate problem for a fixed batch time of one hour. Depending on the problem, one may expect to find these zero reflux holdup effects for easy separations. We have encountered similar profiles when solving Profit Maximization problems (Logsdon et al., 1990), where we considered modified short-cut models which allowed for holdup. As shown in Figure 5, the no holdup short-cut model does not predict the zero reflux policy.

We summarize the performance of our algorithm in Table 2 for the stage-to-stage column model. To evaluate the solution accuracy, one can examine the error residuals for the overall material balance and the first component. Except for the steepest (and smallest) elements near the ends, the approximation errors in the profiles are about 10^{-4} or less.

4.3 Comparison with Constant and Piecewise Constant Reflux Policies

Finally, we compare our results with other conventional reflux policies including, constant optimal reflux, optimal piecewise constant and constant overhead policies, in order to assess their influence when condenser and tray holdup dynamics are modeled. From this comparison we emphasize that recent work with tray to tray holdup models should also be examined carefully, as the effect of holdup could be ignored by considering too simple a reflux policy. Here we resolve the endpoint control problem with an optimal constant reflux policy. This is accomplished by constraining the reflux variable to be the same at all collocation points. The solution is shown in Figure 7. Note here that the constant reflux policy causes the column purity to steadily decline. Obviously, since the reflux policy is not allowed to vary with time, the advantages of a zero reflux policy cannot be considered here.

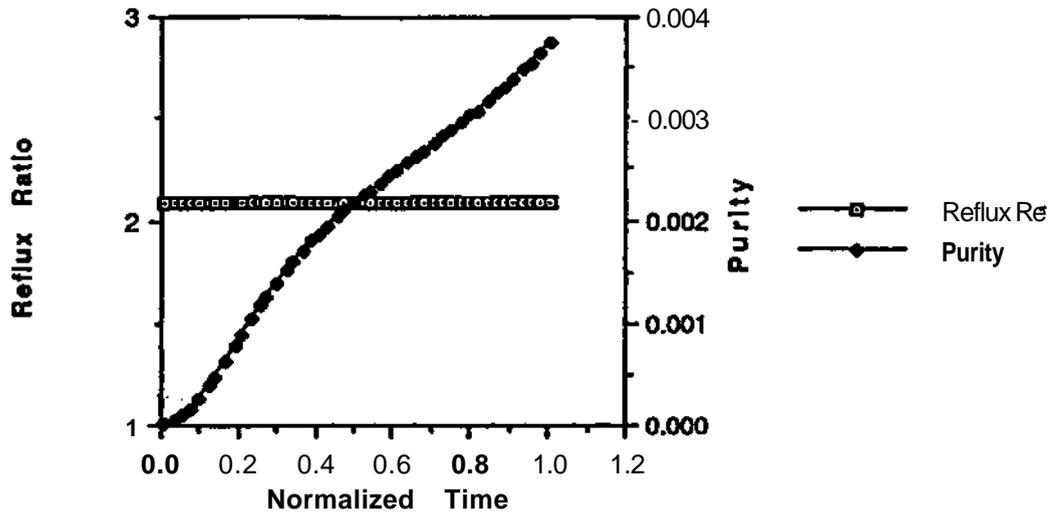


Figure 7: Optimal Constant Reflux Policy and Purity (1 - xd) Profiles with an Endpoint Constraint The open squares indicate the reflux policy (left axis), the closed diamonds indicate the purity profile.

Next we consider a reflux policy obtained through parameterization of the control profile. This approach is similar to the one taken by Mujtaba and Macchietto (1988). Again, this problem formulation is accomplished by requiring the reflux control variable to be the same at each of the collocation points *within each of the variable length finite elements*. The optimal policy for this case is shown in Figure 8 along with the purity profile.

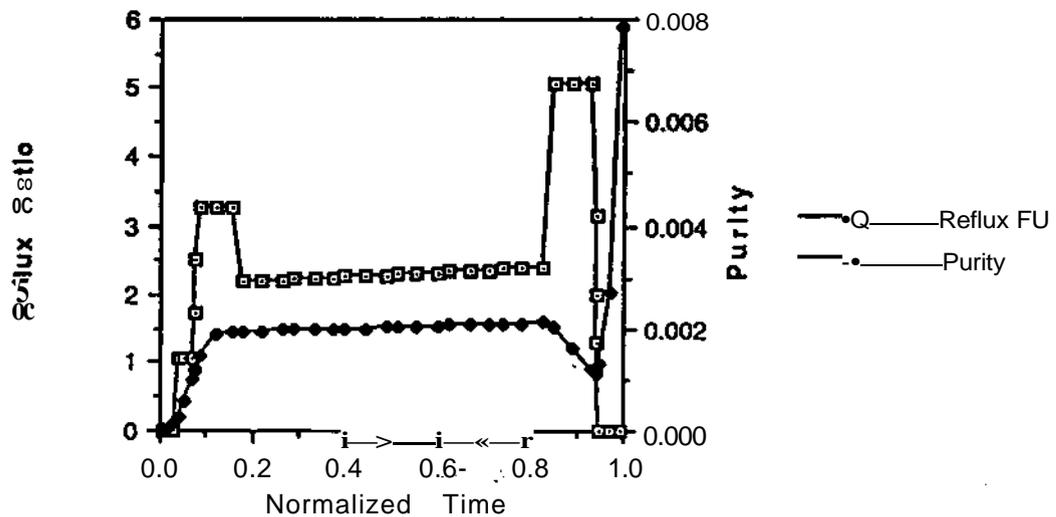


Figure 8: Optimal Parameterized Reflux Policy and Purity ($1 - x_d$) Profiles with an Endpoint Constraint. The open squares indicate the reflux policy (left axis), the closed diamonds indicate the purity profile.

Here we note that the zero reflux policy is also predicted for the parameterization, as in Figures 5 and 6. In fact, one can view the parameterized policy as an approximation to the previous ones. Nevertheless, care should be taken when using a differential equation solver independent of the optimization of the reflux policy since a zero reflux policy may not appear if the switching points are located too far apart. Therefore, small switching times at the initial and final portion of the distillation period may not be detected in this problem formulation. The amount of distillate collected for each of the policies, along with CPU times for the optimizations, is summarized in Table 2.

Finally, we consider a constant overhead policy for this problem. However, since the column is initialized at total reflux with a purity higher than the overhead specification (0.998), we require a short period for the purity to drop to this level (Diwekar and Madhavan (1991) suggest a zero reflux policy for this period.). This is precisely the optimal policy for the path constrained problem that we considered above and we use this as a basis for comparison. From Figures 9 and 10 we see that except for the final "holdup dumping" portion in the endpoint constrained policy, the two cases produce similar profiles. However, as seen in Table 2, the distillate recovered is 9% higher for the endpoint constrained problem and this can be attributed directly to holdup dumping and the zero reflux policy at the end. Thus for this particular case we see that zero reflux policies produce substantial improvements in distillate recoveries. Moreover, from Table 2 we see that the shortcut policy and the constant reflux policy do not allow for a zero reflux and lead to decreases in recovery up to 12 percent

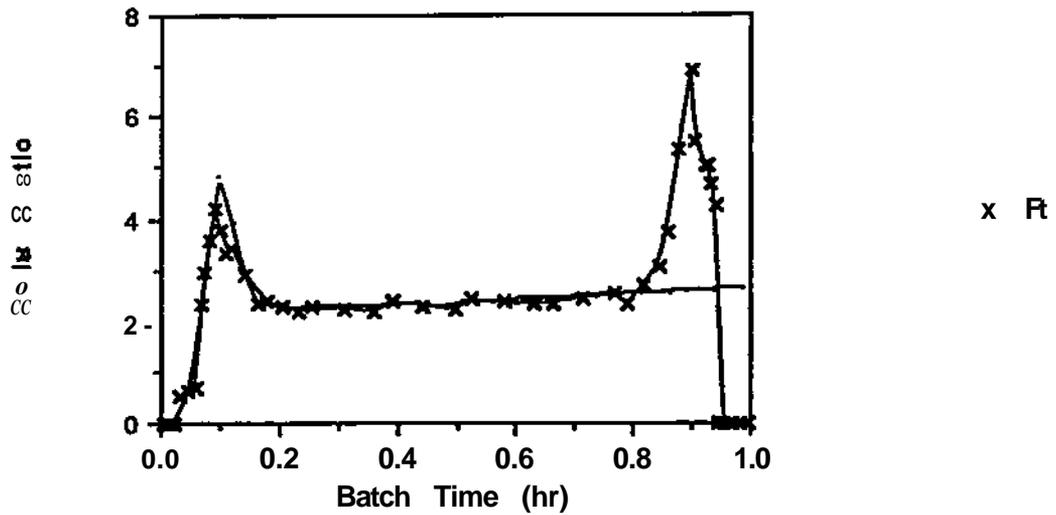


Figure 9: Comparison of Optimal Reflux Policies: Path and Endpoint Constraints(x).

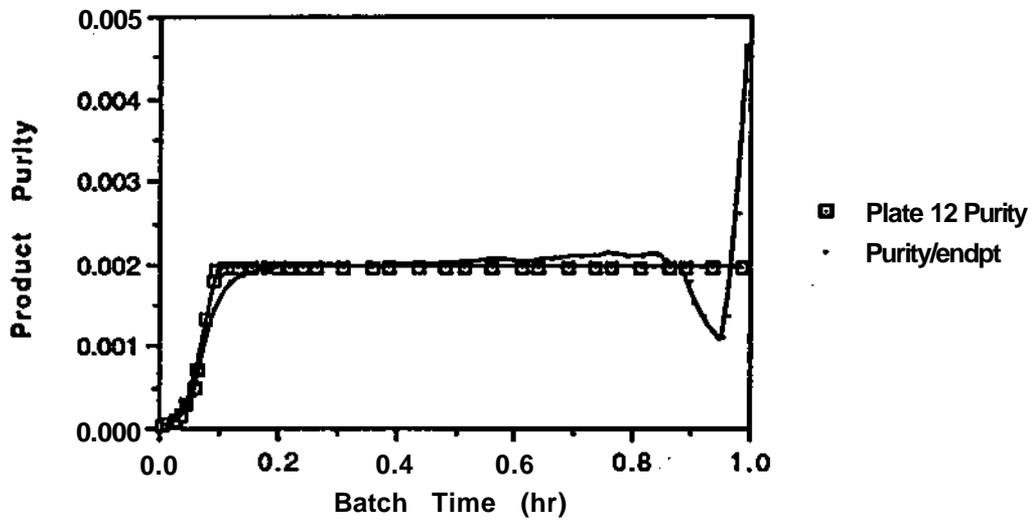


Figure 10: Comparison of Distillate Purities: Path and Endpoint Constraints (open squares). Note that the equivalent distillate purity is 0.998 for a top plate purity of 0.995.

Table 2: Amount of Distillate collected for Various Reflux Policy. Maximum Distillate, Endpoint Constrained Control Problem.

Control Policy	Objective Function	Iterations	CPU Time (VS 3200)
Optimal Control	42.338 (moles)	52	1:57:30
Short Cut	37.029 (moles)	34	0:02:12
Optimal Constant Reflux	38.899 (moles)	6	0:09:53
Parameterized Reflux	42.259 (moles)	51	1:51:20
Path Constrained	38.615 (moles)	51	1:09:44

4.4. Relationship to More General Formulations

The maximum distillate optimization can also be generalized to cover a number of more complex batch optimization problems. For example, consider the boilup rate, $V(t)$, as an additional control profile. Logsdon et al. (1990) and Mujtaba and Macchietto (1991) note that for maximum distillate problems the optimal boilup rate is always constrained at its upper bound. Moreover, Logsdon et al (1990) show that this property is also true for the maximum profit problem as long as the optimal solution shows a positive profit

In addition, note that the optimal reflux policies actually scale with time and inversely with the boilup rate. This can be seen by normalizing time by $x = t/t_f$ and rewriting the DAE model in (9) in terms of t . Note that the algebraic expressions remain unchanged and are written for normalized profiles $R(x)$, $D(x)$, $x(x)$, etc. When $V(x)$ remains at its upper bound the equations can be redefined for a new variable $G3 = V t_f$. For a given $G5$ the normalized profiles remain the same regardless of the final time or boilup rates. Thus for binary separations, optimal profiles need only be generated for different values of purity, initial conditions and values for $G5$.

Finally, we note that the maximum distillation problem is often a subset of more complex batch optimization formulations. First, Coward (1967), Murty et al. (1980) and Mujtaba and Macchietto (1991) related the maximum distillate problem and the minimum time problem for binary systems. On the other hand, for the maximum profit problem with an objective given, for example, by:

$$\text{Max}_{R(x), N, V} [C_d D_{C=1} - C_s S(x=0)] / (t_f + T) - \text{Op}(V) - \text{Cap}(N, V)$$

(where T represents a setup time, N is the number of trays and Op and Cap refer to operating and capital costs respectively), Diwekar (1992) notes that this objective can be separated into:

$$\text{Max}_{N, V} [C_d \{\text{Max}_{R(x)} D(x=1)\} - C_s S(x=0)] / (t_f + T) - \text{Op}(V) - \text{Cap}(N, V)$$

and the maximum distillate policies can be solved in advance and recalled when needed. In this way the more difficult part of a batch process design can be solved for a normalized problem "off-line" (for a fixed N and initial conditions) and can be largely decoupled from other aspects of the batch process design or operating schedule.

Of course, these observations only apply to simple batch distillation operating policies. More work needs to be done in order to consider more general constraints on optimal profiles, more complex operating policies such as the recycling of intermediates, accommodating multiple cuts and offcuts, and the incorporation of dynamic behavior into scheduling and planning activities. Mujtaba and Macchietto (1988, 1992) and Farhat et al. (1990) describe problem formulations for these cases. Future work will incorporate the more complex reflux policies described here into these problem formulations.

5. Conclusions

Batch distillation remains an important task for a wide variety of chemical processes. While many studies have considered various aspects of distillation optimization, the influence of the column holdups on the optimal operating policy remains a challenging problem. In this study we predict unusual and advantageous optimal reflux policies that exploit holdups in batch columns. To illustrate these policies, we consider two cases for the maximum distillate problem which differ in the specification of the purity constraint. The path constrained case was solved with the purity constraint written for the top plate instead of the distillate in order to keep the problem at index 2; the endpoint constrained problem avoids this difficulty. Both cases were solved through application of orthogonal collocation on finite elements and optimization of the resulting algebraic system with a tailored version of Successive Quadratic Programming (SQP). For these problems, we did not need to

enforce the residual constraints for the approximation error directly. Instead the elements were simply allowed to vary between upper and lower bounds. Monitoring the residuals at the optimum showed that the resulting profiles had approximation errors less than 10^{-5} .

The optimal profiles for both of these maximum distillate cases reveal some interesting characteristics. For the path constrained problem, the optimal reflux policy corresponds to zero reflux (maximum take-off) until the path constraint becomes active. At this point the policy corresponds to constant overhead for the remainder of the operation. The endpoint constraint, on the other hand, introduces another characteristic. While it is similar to the path constrained policy at the beginning, it also contains a zero reflux portion at the end. At this point the contents of the holdup are simply "dumped" in order to maximize the distillate. Due to this effect, an increase of 9% is realized in the final distillate. In addition, we compare these two cases with more conventional operating policies. First, the path constrained case serves as a reasonable approximation to the constant overhead policy. An optimal piecewise constant reflux policy simply approximates our optimal endpoint constrained policy and also contains zero reflux portions (provided that piecewise constant portions are small enough). Moreover, optimal policies based on shortcut models without holdups or constant reflux do not contain zero reflux portions and consequently are suboptimal. The results of these policies are up to 12% less than the optimal endpoint constrained case. Finally, we summarize the computational resources required for these optimizations and note that constrained nonlinear programs of up to 8000 variables are solved in under two CPU hours (Vaxstation 3200) with our approach.

In addition, our method can be applied to multicomponent systems to exploit column dynamics (e.g. initialization and holdup effects) for maximum recovery of distillate product. Diwekar (1992) notes that the maximum distillate problem is an important subproblem for maximum profit optimization. Moreover, Macchietto and Mujtaba (1992) show that more complex batch optimizations, involving multiple product cuts and offcuts, can be handled by solving maximum distillate problems within an inner loop.

Acknowledgments

Financial support from the Engineering Design Research Center, an NSF supported Engineering Center at Carnegie Mellon University, is gratefully acknowledged. The second author is also grateful to the Mathematics and Computer Science Division of Argonne National Laboratory where the later stages of this work were supported in part by the

Applied Mathematical Sciences subprogram of the Office of Energy Research, U. S. Department of Energy, under Contract W-31-109-Eng-38.

References

1. Al-Tuwaim, M. S. and W. L. Luyben, / & *EC Research*, 30, p. 507 (1991)
2. Converse, A. O. and G. D. Gross ,*Ind. Eng. Chem. Fund.*, Vol. 2, No. 3, p. 217 (1963)
3. Coward, I., *Chem. Eng. Sci.*, 22,p. 503, and p. 1881 (1967)
4. Grille, PJ2. and G.V. Reklaitis, *Computers and Chemical Engineering*, Vol. 10, No. 4, pp. 389 (1986)
5. Cuthrell, J. E., and L.T. Biegler, *Computers and Chemical Engineering*, Vol. 9, No. 3, pp. 257 (1985)
6. Cuthrell, J. E., and L.T. Biegler, *AIChE J.* 33 , pp. 1257 -1270 (1987)
7. Cuthrell, J. E. and L.T. Biegler, *Computers and Chemical Engineering*, Vol. 13, No. 1/2, pp. 49-62 (1989)
8. Diwekar, U. M., "A Unified, Efficient Approach to Solving Simultaneous Optimization and Optimal Control Problems in Batch Distillation," *submitted to AIChE J.* (1992)
9. Diwekar, U. M, PhD Thesis, IIT Bombay (1988)
10. Diwekar, U. M., Malik, R.K., and K.P. Madhavan, *Computers & Chemical Engineering* Vol. 11, No. 6, p.629 (1987)
11. Diwekar, U. M. and K.P. Madhavan, *Computers & Chemical Engineering*, Vol. 15, No. 12, p.833 (1991)
12. Domenech, S. and M. Enjalbert , *Chem. Engng Sci.*, Wol 29, p. 1519 (1974)
13. Domenech, S. and M. Enjalbert , *Computers & Chemical Engineering* ,Vol. 5, p. 181 (1981)
14. Eaton, J.W., J.B. Rawlings and T.F. Edgar, " Model Predictive Control and Sensitivity Analysis for Constrained Nonlinear Processes," *IFAC Workshop on Model Based Control*, Atlanta, GA (1988)
15. Farhat, S., M. Czernicki, L. Piboleau, S. Domenech, *AIChE J.*, 36,9, p. 1349 (1990)
16. Hansen, T. T., and S. B. Jorgensen, *The Chemical Engineering Journal*, 33, p. 151 (1986)
17. Kerkhof, L. H. J. and H. J. M. Vissers , *Chem. Eng. Sci.*, Vol. 33, p. 961 (1978)

18. Logsdon, J.S. and L.T. Biegler, *Ind. & Eng. Chem. Res.*, Vol. 28, No. 11, p. 1628(1989)
19. Logsdon, J.S., U.M. Diwekar and L.T. Biegler, *Chemical Engineering Research and Design*, **68**, p. 434 (1990)
20. Logsdon, J. S. and L. T. Biegler, *Chem. Engr. Sci.*, 47,4, p. 851 (1992)
21. **Luyben, W. L., *Ind. Eng. Chem. Process Des. Develop.*, **10**, p. 54 (1971)**
22. Luyben, W. L., *I & EC Research*, 27, p. 642 (1988)
23. Mayur, D. N. and R. Jackson, *Chem. Eng. J.*, Vol. 2, p. 150 (1971)
24. **Mujtaba, I. M. and S. Macchieto, *Proceedings 12th IMACS World Congress*, Paris, July 18-22 (1988)**
25. **Mujtaba, I. M. and S. Macchieto, *SIMO'88 - Simulation et optimisation en Genie des Procedes*, Toulouse (France), Sept. 14-15 (1988)**
26. **Mujtaba, I. M. and S. Macchietto, *Proc. Process Systems Engineering '91*, Paper 1.19 (1991)**
27. Mujtaba, I. M. and S. Macchietto, Paper 61c, presented at National AIChE Meeting, New Orleans, Spring, 1992
28. Murty, B. S. N., K. Gangiah and A. Husain, *The Chemical Engineering Journal*, 19, p 201 (1980)
29. Pigford, R. L., Tepe, J. B., and C. J. Garrahan, *Ind. Eng. Chem.*, 43, p. 2592 (1951)
30. Reid, R. C, J. M. Prausnitz and T. K. Sherwood, **Properties of Gases and Liquids**, Me Graw-Hill, New York (1977)
31. Renfro, J.G., Morshedi, A.M., and O. A. Asbjomsen, *Computers and Chemical Engineering*, Vol. 11, No. 5 , pp. 503- 517 (1987)
32. Robinson, E. R., *Chem. Eng. Sci.*, 25, p. 921 (1970)
33. Vasantharajan, S. and L.T. Biegler, *Computers and Chemical Engineering*, Vol. 12, No. 11, p. 1089 (1988)
34. Vasantharajan, S., J. Viswanathan and L.T. Biegler, *Comp. Chem. Engr.*, 14, 8, p. 907 (1990)