OPTIMAL CONTROL AND ON-LINE OPERATION
OF REACTIVE BATCH DISTILLATION

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ABSTRACT

While both the dynamic modelling, and to a lesser extent, the optimisation of reactive batch distillation have been studied in the past, issues related to the control and on-line operation of such a process have not been properly addressed. In this study a-priori optimal profiles of the operating variables are established for an industrial reactive distillation by maximising the profitability of the process subject to constraints on the reboiler temperature and the accumulator composition. The control properties at optimal and non-optimal conditions are assessed. A PI-controller is designed to implement the optimal profiles and the controller performance is tested using a simulation model including disturbances in the reaction model and reboiler heat duty. The fully automatic operation of the process, according to the optimal policies, is demonstrated in conjunction with a real time control system.

INTRODUCTION

Batch distillation is used in the chemical industry for the production of small amounts of products with high added value and for processes where flexibility is needed. A chemical reaction and multi-stage distillation can be carried out simultaneously. This combined unit operation, called reactive batch distillation, especially suits those chemical reactions where reaction equilibrium limits the conversion in a normal reactor to a low-to-moderate level. By continuously separating low-boiling products from reactants and other products while the reaction is in progress, the reaction can proceed to a much higher level of conversion. Also, the removal of a light product results in a decrease in its concentration in the liquid phase thereby increasing the liquid temperature and the reaction rate.

While both the dynamic modelling of reactive batch distillation, and to a lesser extent, its optimisation has been studied in the past (Egly *et al.* 1979, Cuille and Reklaitis, 1986, Albet *et al.*, 1991, Mujtaba and Macchietto, 1992), issues related to the controllability and control of such a process have not been properly addressed. Some attention to the implementation of optimal control profiles for batch distillation has been given by Quintero-Marmol and Luyben (1992) and Bosley and Edgar (1992). So far, only Sørensen and Skogestad (1992) have considered the controllability of reactive batch distillation columns.

This study addresses several aspects of the optimal control of a reactive batch distillation process with reference to a specific industrial application. The control objective is to maximise the profitability given constraints on the maximum allowable reboiler temperature to avoid thermal decomposition of the product and minimum loss of volatile reactant in the distillate. Manipulated variables for control are the heat input to the reactor, Q_R, and the internal reflux ratio, $R = L/V$. It is assumed that the condenser duty, Q_C, is used for pressure control and the distillate flow, D, for condenser level control. Of particular interest are i) establishing a-priori optimal profiles of the operating variables, ii) assessing the controllability properties at

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the optimal conditions, iii) the design of controllers to implement the optimal profiles and iv) the verification of stability and control performance of the controller.

REACTIVE BATCH DISTILLATION MODEL

The model used in this study is valid under the following assumptions: A1) A staged model is used for the distillation column with the trays numbered from the top down (accumulator=A, condenser j=1, reboiler j=N), A2) a multicomponent mixture is considered, A3) perfect mixing and equilibrium between vapour and liquid on all stages, A4) the vapour phase holdup is negligible compared to the liquid phase holdup, A5) constant stage pressures and plate efficiencies, A6) constant molar holdups on the plates and in the condenser, A7) fast energy dynamics, A8) total condensation with no subcooling in the condenser, A9) the chemical reaction is limited to the reboiler and A10) Raoult's law for the vapour-liquid equilibrium. This simple model was used for the open-loop optimisation while a more complex model including linear tray hydraulics was used for simulations and control studies. The linear hydraulics model has the form $L_j = L_{0,j} + (H_j - H_{0,j})/\tau$, where $L_{0,j}$, L_j, $H_{0,j}$ and H_j are the initial and current instant liquid flow rate from and molar holdup on the trays, respectively, and τ is the hydraulic time constant.

The reaction considered is a condensation polymerisation reaction between a dibasic aromatic acid (R_1: Terephthalic acid) and two glycols (R_2: 1,2-Propanediol and R_3: 1,6-Hexanediol) which forms a polymer (P: Poly(propylene/hexylene)-terephthalate) and water (W). The polymer forms a separate, solid phase and is therefore not included in the distillation model. High conversion is achieved by removing the water in the distillate. The physical properties data for the liquid components were taken from Coulson and Richardson (1983) for water and 1,2-propanediol, and from Daubert and Danner (1985) for 1,6-hexanediol and terephthalic acid. The data for the process used in the optimisation and the simulations are given in Table 1.

Column:	reboiler + 5 trays + total condenser + accumulator
Reaction:	$1.0\ R_1 + 0.7\ R_2 + 0.3\ R_3 \leftrightarrow P(s) + 2.0W$
Reaction rate:	$r = r_{R1} = k_1 c_{R1}(c_{R2} + c_{R3}) - k_2 c_W$
Rate constants:	$k_i = k_{0i} \exp\left(-\frac{E_i}{R} * \left(\frac{1}{T_N} - \frac{1}{T_0}\right)\right)$
Activation energies:	$E_1 = E_2 = 18\ \text{kcal/mol} = 7.5 \cdot 10^4\ \text{kJ/kmol}$
Frequency factors:	$k_{1,0} = 0.03$ and $k_{2,0} = 0.25 \cdot k_{1,0} = 0.0075$
Reference temperature:	$T_0 = 500.0$ K
Pressure in column and reactor:	1 atm
Hydraulic time constant, τ:	0.0018 h = 6.5 s
Initial holdups (reboiler, trays, condenser, accumulator):	20.6, 0.1, 1.7 and 10^{-6} kmol
Initial charge composition j=N:	$x^0(W, R_2, R_3, R_1) = [0.0097, 0.3444, 0.1505, 0.4854]$
Initial tray compositions j=1,N-1:	$x^0(W, R_2, R_3, R_1) = [0.999999, 0.000001, 0.0, 0.0]$
Value of product P:	$C_{prod} = 50$ \$/kmol
Cost of steam:	$C_{heat} = 3\ \text{\$}/10^6\ BTU = 2.844 \cdot 10^{-6}$ \$/kJ
Total reaction time:	specified or optimised, hr ; range=[0,24.0]
Reboiler heat duty, Q_R:	specified or optimised, kJ/hr ; range=[0.7,0.9]$\cdot 10^6$
Reflux ratio, R:	specified or optimised ; range=[0.9,1.0]

Table 1: Process data for optimisation and simulation.

OPTIMAL A-PRIORI OPERATION

The optimal reflux ratio and reboiler heat duty are found based on some optimisation criterion. In this study the optimisation problem has been solved according to a maximum profit control objective. In general, the optimal control problem can be stated as (Vassiliadis, 1992):

$$\min_{\dot{x}(\cdot), x(\cdot), y(\cdot), u(\cdot), v, x(t0), t_f} \Phi(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f) \qquad (1)$$

subject to

$$f(\dot{x}(t), x(t), y(t), u(t), v, t_f, t) = 0; \; h_1(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f) = 0; \; h_2(\dot{x}(t_f), x(t_f), y(t_f), u(t_f), v, t_f) \geq 0$$

In addition there are upper and lower bounds on the state (x, y), design (v) and control (u) variables and also on the time (t_f). The optimisation of the DAE optimisation problem is solved by parameterisation of the control vector into a finite number of variables which are optimised using a standard nonlinear optimisation technique. The method includes two levels, the first level performs an integration of the differential and algebraic equations for fixed values of the decision variables, the second level optimises those decision variables and satisfies end point constraints. A complete solution of the DAEs for each trial value of the decision variables is required. The optimal operating policy is determined here by optimising the overall profit [$/hr]:

$$\max_{R, Q_R} P^{\$} = \frac{1}{t_f} \left(C_{prod} A_{prod} - C_{heat} \int_0^{t_f} Q_R(t_f) dt \right) - C_{fixed} \qquad (2)$$

where C_{prod} is the value of product P ($/kmol\;monomer\;block$), A_{prod} is the amount of product P at t_f ($kmol$), C_{heat} is the cost of steam to the reboiler ($/kJ$), t_f is the final time (hr), C_{fixed} are the fixed costs ($/hr$) and $P^{\$}$ is the profitability ($/hr$). Zero cost of the raw material is assumed. Note that since the initial charge is fixed and we assume all unreacted feed is recycled to the next batch at no cost, this will not affect the location of the optimum. We use $C_{fixed} = 0$ since the optimum will also be independent of the fixed costs. A single discretisation interval is used with constant controls resulting in two variables (R and Q_R) for the optimisation. The constraints are 1) Negligible loss of reactant R_2 in the accumulated distillate ($x_{A,W}(t_f) \geq 0.999$) and 2) reboiler temperature below $240^{\circ}C = 513.15\;K$ to avoid thermal decomposition of the end-product ($T_N(t_f) \leq 513.15$). All simulations have shown that the reboiler temperature is highest at the end of the reaction, so keeping the endpoint temperature constraint is sufficient. If this were not the case, the constraint on the reboiler temperature could be added as a path constraint instead of as an end-point constraint. With the process data given in Table 1 the optimal values are $R = 0.950$ and $Q_R = 0.747 \cdot 10^6$ kJ/hr with $t_f = 22.1$ hr, which yields $x_{A,W}(t_f) = 0.9997$, $T_N(t_f) = 494.4$ K and $\chi_{R1}(t_f) = 0.97$ where χ_{R1} is the conversion of reactant R_1. Solutions with different values of the monomer product, e.g. $C_{prod} = 100$ $/kmol or $C_{prod} = 25$ $/kmol, give almost the same optimal results. The temperature profiles for the process given the optimal control values are given in Fig. 1.a. The reboiler temperature is more or less
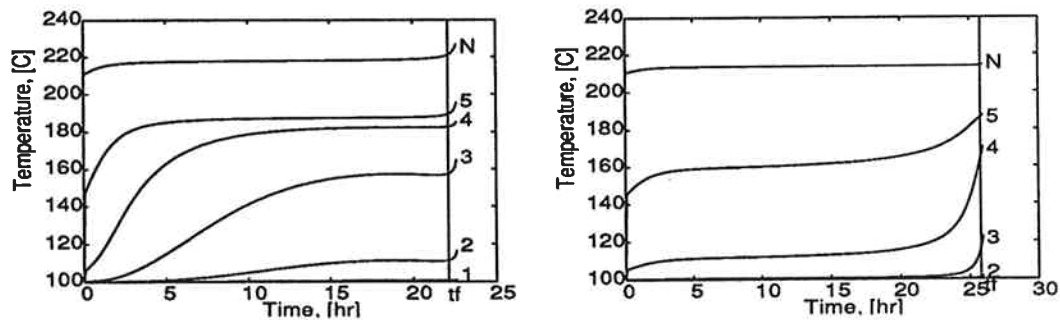

Figure 1: Temperature profiles for a) optimal and b) sub-optimal conditions.

constant during the whole batch but the temperature in the top section of the column increase slowly. There is a sudden increase in temperature on all trays immediately after the optimal final time. The end of the reaction in the real plant can therefore be easily detected using temperature measurements on any one of the trays or in the reboiler. Also note that the optimum with respect to time is very flat. Indeed, ending the batch after 16 hr (25 % time saving) still gives 99 % of the optimal profitability with a conversion of R_1 of 0.71. The reason for this insensitivity to the value of t_f is that we have assumed that there is no cost involved in recycling the unreacted feed.

When using sub-optimal values for R and Q_R (e.g. $R = 0.954$ and $Q_R = 0.691 \cdot 10^6$ kJ/hr) a different response is obtained (Fig. 1.b). The constraints are still satisfied but the previous optimal conversion is only reached at t=25.8 hr. The profitability is 15 % lower, at 16.9 $/hr. The temperatures for this sub-optimal operation are lower than under optimal conditions showing that we are over-refluxing the column. Also, detection of the termination conditions based on temperature alone is much more difficult.
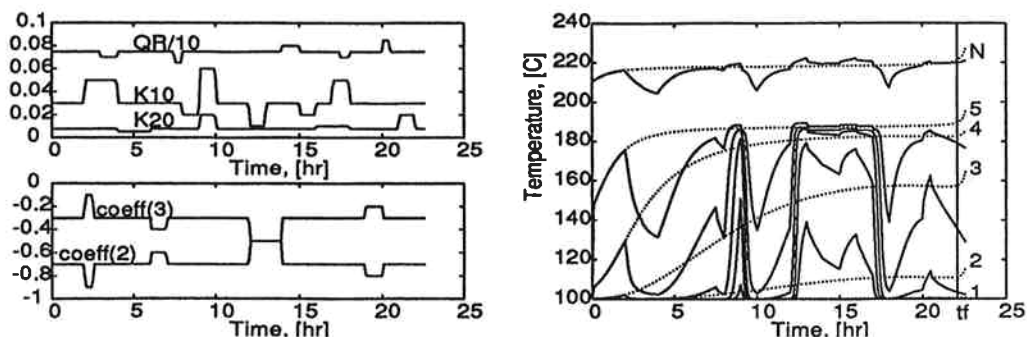
Figure 2: a) Disturbances in the process and b) temperature profiles for the uncontrolled process.

CONTROL ASPECTS

Since the formation of product resembles a continuous feed to the reboiler (Sørensen and Skogestad, 1992) the issues involved in control of a reactive batch column are more similar to conventional continuous distillation control than to batch distillation control. In a conventional batch column one may usually implement the optimal values of R and Q_R in an "uncontrolled" open-loop fashion. However, for reactive distillation the operation is usually much more sensitive to the exact value of the reflux ratio R. A slightly too low R may result in a distillate flow which is larger than the amount of light component (W) formed in the reaction, and the difference must be provided by loss of the intermediate boiling reactant (R_2). Therefore there is a need to implement the optimal policy in some feedback fashion which maintains control of the composition of the distillate. We propose to keep Q_R constant at its optimal value and adjust R to control the temperature on some tray inside the column. The temperature measurement should be placed on a tray with a large response to changes in the controller input. By controlling the temperature on one of the trays one will also to some extent indirectly control the reactor temperature since there is a strong coupling between the compositions in the column (Sørensen and Skogestad, 1992).

ONE-POINT COLUMN CONTROL

We now consider implementing the optimal solution. Various disturbances are introduced in the simulation model to test the sensitivity to process changes (Fig. 2.a). The disturbances are assumed to be in 1) the reboiler heat duty Q_R, 2) the frequency factors for the forward and reverse reaction, k_{10} and k_{20} and 3) the stoichiometric coefficients of the alcohols (reactant 2 and 3) in the rate equation. Disturbances 2 and 3 are used to represent some uncertainty in the reaction mechanism. First consider the uncontrolled case where R is kept constant. The temperature profiles (Fig. 2.b solid lines) deviates significantly from the optimal profiles (Fig. 2.b dotted lines). The disturbances yield breakthrough of the intermediate component resulting in some loss of reactant R_2 ($x_{A,W} = 0.989$ for the accumulated distillate) and the batch therefore does not satisfy the specifications. Also the reboiler temperature deviates up to 12° C from the optimal one which may result in a varying quality of the product P between batches.
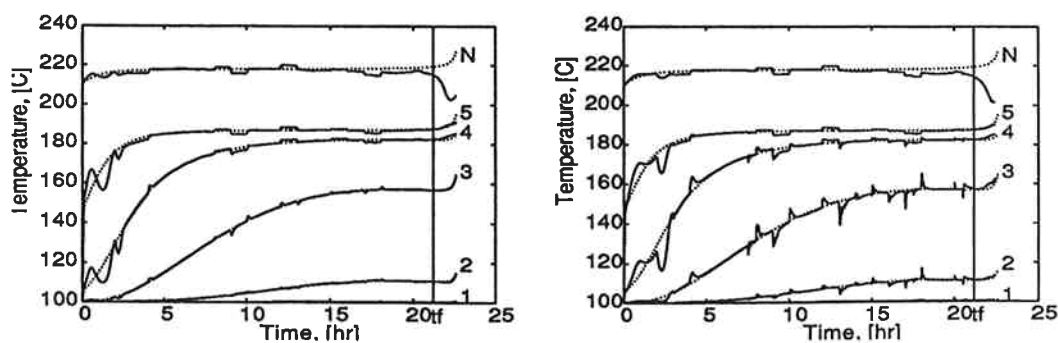

Figure 3: Temperature profiles using a) continuous controller and b) real time control system.

Next consider implementing the optimal policy in a feedback fashion by varying R to keep a selected tray

temperature constant. The temperature measurement is placed on tray 3. Reflux ratio R is the manipulated variable and Q_R is kept at its optimal value. The set point for the temperature controller is the optimal temperature profile for tray 3 as a function of time (fitted to a polynomial). A time delay of 15 seconds was added to the temperature measurement and the controller settings used are $K_c = -0.01$ and $\tau_i = 0.1$. The controller is tuned using a linearised model at given operating points. As can be seen from the results in Fig. 3.a. (solid lines) good control is achieved. The deviation in the reboiler temperature from the optimal profile (dotted lines) is small during most of the batch, but increases up to 20° C towards the end. The optimal conversion ($\chi_{R1} = 0.97$) is reached at $t = 21.2$ hr. After this time the reboiler temperature starts drifting away from the optimal value. For the controlled process the reboiler temperature is decreasing instead of increasing as for the optimal process. This is because the controller maintains the temperature on tray 3 at its set point as given by the optimal profile. The uncontrolled temperature on tray 3 increases with time as the reaction proceeds. But due to the disturbances the reaction rate is quicker than under the optimal conditions. Keeping the temperature at the optimal set point will therefore hold light component back in the lower part of the column causing the reboiler temperature to decrease. Detecting the end of the reaction based on the criteria from the optimal temperature profile, that the reaction is ended when the temperatures start rising rapidly, can not be applied in this case. However, the fact that the reboiler temperature is drifting away from the optimal in either direction is an indication that the reaction is ended. The end point can therefore be estimated to when the reboiler temperature deviates from the optimal on either side by an amount greater than e.g. 5° C.

The process was then interfaced to an industrial real-time control system, Paragon. All controllers were implemented using Paragon's own facilities. Instead of using a polynomial to describe the simulation temperature set point profile only 16 measurement points were used. The sampling time was 1 minute. As shown in Fig. 3.b good control is achieved and the responses are very similar to those obtained with the simulated continuous controller.

CONCLUSIONS

The optimal operating values for the reflux ratio and reboiler heat duty for a batch distillation column with reaction in the reboiler have been determined according to the maximum profit control objective. It has been found that large time savings can be made with only small reductions in profitability. The optimal policy should be implemented using feedback control to avoid loss of reactant ("breakthrough") in the distillate waste product. We select to control an intermediate column temperature which location is selected based on a sensitivity analysis of the optimal temperature profiles. The column has been controlled using a simple continuous PI-controller and with a real time control system with disturbances in the reaction parameters and in the reboiler heat duty. Good performance has been achieved in both cases. For this industrial application it has been found that the end of the reaction can be detected based on temperature measurements in the reboiler. An alternative to feedback control is over-refluxing but this may lead to significant cost penalties.

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