PROCESS DESIGN AND CONTROL OF A REACTIVE DISTILLATION SYSTEM

Pinky Panjwani, Myrian Schenk[1], Michael C. Georgiadis and Efstratios N. Pistikopoulos

Imperial College London,
Department of Chemical Engineering, Centre for Process Systems Engineering, London SW7 2AZ, UK

Abstract: A Mixed-Integer Dynamic Optimization (MIDO) model and solution strategy to study the interactions between process design and process control/operability of an ethyl acetate reactive distillation system is presented. It is shown that the resulting design and control scheme can guarantee feasible operation under bounded uncertainty at a minimum total average cost, representing 17 to 20% savings over the original design. *Copyright © 2004 IFAC*

Keywords: Process design; Control; Reactive distillation; Mixed integer optimization.

1. INTRODUCTION

In recent years chemical process industries have shown an increasing interest in the development of reactive distillation processes combining reaction and separation mechanisms into a single, integrated unit. Such processes offer several important advantages such as reduction in total costs and energy consumption, overcoming of thermodynamic limitations, (e.g. azeotropes) and increased reaction yield and selectivity. However, reactive distillation is not extensively used in the chemical process industries since its operation is more difficult and poses higher requirements on the quality of the design and control than conventional flowsheets in which a reactor is typically followed by a train of distillation columns (Engell and Fernholz, 2003).

Modelling of reactive distillation has received considerable attention over the last 10 years and several key contributions have appeared in the literature (see the book of Doherty and Malone, 2001 and the overview of Noeres *et al*., 2003). Pilavachi *et al.* (1997) presented an extensive discussion of several important aspects that affect the accurate modelling of reactive distillation processes. Schenk *et al.* (1999) described in considerable detail a hybrid-modelling environment in which a reactive distillation process can be simulated using a combination of equilibrium and mass transfer models, both in steady state and dynamic modes.

The control of reactive distillation has received some attention in recent years. Sneesby *et al.* (1999) discussed the advantages of a combined composition and conversion scheme for an ETBE column. Monroy-Loperena *et al.* (2000) studied the control problem of an ethyl glycol reactive distillation system and proposed a robust PI control configuration. Sneesby *et al.* (2000) proposed an integrated control scheme for an ETBE column by permitting the control objectives to be changed on-line in order to reflect changing economic contracts. Vora and Daoutidis (2001) studied the dynamics and control of an ethyl acetate reactive distillation system and proposed a new feed configuration for the two reactants that allows higher conversion and purity than the conventional configuration, which involves feeding in a single tray. Al-Arfaj and Luyben (2002) compared the control of an ideal reactive distillation column with that of a similar real chemical system, the production of methyl acetate. A number of control structures were evaluated for both systems. Gruner *et al.* (2003) proposed a non-linear control scheme for an industrial reactive distillation column operated by Bayer AG. Bisowarno *et al.* (2003) investigated model gain scheduling for an ETBE reactive distillation column. It was illustrated that the proposed control strategy outperforms the standard PI control.

The work in the area of integration of design and control in reactive distillation systems is very limited. Heath *et al.* (2000) studied the interactions of design

[1] Present address, Air Products PLC, Molesey Road, Walton-on-Thames, Surrey, U.K.

and control in an ethyl glycol reactive distillation system assuming that the process structure is fixed (e.g. number of trays, feed tray location, etc). Schenk (1999) and Georgiadis *et al.* (2002) compared sequential and simultaneous approaches to design and control of a reactive distillation system, using advanced optimization techniques. Again the process and control structure were assumed fixed.

The aim of this work is to explore the synergistic benefits between process design, process control and operability in a reactive distillation column for the production of ethyl acetate. The process structure (number of trays, feed tray location), the process design (column diameter, reboiler and condenser surface areas) and control structure/design (pairings of measurements and manipulated variables, controller tuning parameters) are all simultaneously optimized. The problem is posed as a Mixed-Integer Dynamic Optimization (MIDO) problem and solved using recent algorithmic advances developed in our group at Imperial College (Bansal, *et al.,* 2003).

2. PROBLEM DEFINITION

The reactive system of ethyl acetate was considered in this work, since experimental data is openly available in the literature. This enables to validate the modelling results as well as the application of the proposed optimization framework.

The main problems encountered in achieving high purity products in the ethyl acetate reactive distillation system are summarized below (Bock *et al*. (1997); Chang and Seader (1988)):

- ✓ unfavourable reactant conversion;
- ✓ similar K-values of ethanol, water and ethyl acetate;
- ✓ temperature profile in the column;
- ✓ the system is strongly non-ideal due to the presence of ethanol, acetic acid and water. The separation of pure components is very difficult due to the existence of five normal azeotropes, namely, ethanol-water; water-acetic acid; ethyl acetate-ethanol; ethyl acetate-water; ethanol-ethyl acetate-water.

Full details of this system as well as a rigorous dynamic model together with the thermodynamic method employed are presented in Georgiadis *et al.* (2002).

Figure 1 depicts the ethyl acetate column superstructure considered where a single feed configuration is assumed. The objective is then to design the column and its required PI control scheme at minimum total annualised cost (capital cost and operating cost), which can guarantee feasible operation over a finite time horizon, subject to a set of constraints (Georgiadis *et al*., 2002) such as:

- ✓ model equations;
- ✓ high-frequency sinusoidal disturbance in the acetic acid inlet composition;
- ✓ slow-moving disturbance in the cooling water inlet temperature representing diurnal, ambient variations;
- ✓ product quality specifications;
- ✓ minimum column diameter to avoid flooding;
- ✓ temperature driving forces in the reboiler and condenser;
- ✓ fractional entrainment limits;
- ✓ control structure specifications.

The solution of this problem would determine:

(a) the optimal process design, i.e., number of trays and feed location (discrete decisions), the diameter of the column, and the surfaces areas of reboiler and condenser (continuous decisions);
(b) the optimal PI control design, i.e., the pairings of manipulated and controlled variables (discrete decisions) and the tuning parameters (gain, integral time) of the PI controllers (continuous decisions).

Note that an original design of diameter 5.2 m, reboiler area 195 m^2 and condenser area 495 m^2 is considered as a base case for comparison of the results obtained in this paper.

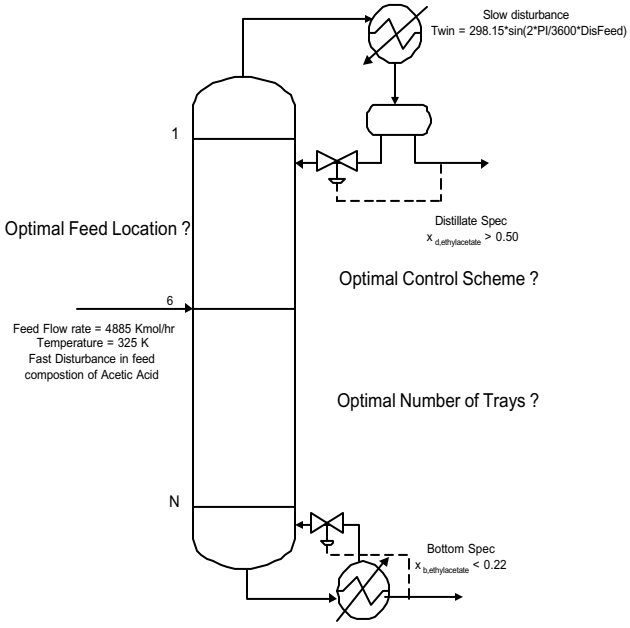

Fig. 1 Ethyl acetate column superstructure

3. SOLUTION STRATEGY

The methodology employed in this work relies on a systematic mixed-integer dynamic optimization (MIDO) framework exploring simultaneously the interactions between process and control system design (Bansal *et al*., 2003). Two strategies were employed. In the first strategy, the control scheme is fixed, while the design of the column is optimized. The column design includes both discrete decisions

such as number of trays and feed location, as well as continuous decisions (column diameter, exchanger areas, controller tuning parameters). The second strategy additionally incorporates the control structure selection (discrete decision). In this way, the economic and operability benefits of simultaneously optimizing the process and control design can be revealed and further assessed. Full details and files can be obtained from the authors by request.

3.1 MIDO Strategy with Fixed Control Structure

The ethyl acetate column superstructure consisted of 13 trays with feed located on the 6th tray. Some restrictions were imposed for the solution of this large-scale problem. Constraints like feed and reflux entering in only one tray; and, feed entering below the reflux were included in the mathematical framework. The fixed control scheme incorporated had two proportional integral controllers, as normally used in distillation systems, namely:

- PI1: Reflux rate (R) – Top ethyl acetate composition ($x_{4,d}$)
- PI2: Steam flow rate (F_s) – Bottom ethyl acetate composition ($x_{4,r}$)

Since the period of the slow-moving disturbance in the cooling water temperature is 3600 hours, it is deemed that this time is adequate as time horizon while rendering the large-scale dynamic optimization problem tractable. The objective function is defined as follows,

$$(Expected)Cost = \min_{d} \left\{ C_{cap} + \sum_{j=1}^{ns} w_j \cdot C_{op,j} \right\} \quad (1)$$

For this particular case, the resulting MIDO problem consists of 1652 model equations (81 differential and 1571 algebraic) with 26 binary (0-1) variables and 36 optimization parameters (9 inequality constraints and 27 equality constraints). The MIDO problem is decomposed into primal problem and master problem. The dynamic optimization primal problems (NLPs) were solved using gPROMS/gOPT (Process Systems Enterprise Ltd., 1999) and the master problems (MILPs) were solved using GAMS/CPLEX. (Brooke *et al.,* 1992).

Final convergence, i.e. the solution of the problem was obtained in two iterations. Table 1 presents the full details of the primal and master solutions for both iterations. The optimal column configuration has 12 trays with the feed located on the 6th tray and the corresponding final total cost is 536125 $/yr.

3.2 Overall MIDO Strategy

The overall MIDO strategy considers the same column superstructure, objective function and optimization variables as the previous strategy but now also includes the control structure selection as additional optimization decisions.

Some process knowledge restrictions were imposed for the solution. In the master problems, based on the fact that a very long time response is undesirable between the action of a manipulated variable and its effect on a controlled variable, pairings such as Reflux rate – Bottom ethyl acetate composition, Distillate flowrate – Bottom ethyl acetate composition, Steam flowrate – Top ethyl acetate composition, Bottoms flowrate – Top ethyl acetate composition, are not considered from the search space; and, due to the available degrees of freedom in the system, only two controllers are allowed.

Table 1 Progress of Iterations for the MIDO strategy with fixed control structure

Iteration No	1	2	3
Primal Solutions			
Discrete Decisions			
No of trays	13	12	11
Feed location	6	6	5
Process Design			
Diameter of column	5.00	4.95	
Reboiler surface area	170.00	168.00	--Inf– (Infeasible point)
Condenser surface area	480.00	478.00	
Controller gains			
PI1	25	25	--Inf–
PI2	5726.00	5725.66	
Controller Reset times			
PI1	60	60	--Inf-
PI2	4936.00	4935.54	
Controller Bias			
PI1			
PI2	3065.00	3074.54	-Inf--
	59000.0	58950.0	
Capital Cost	297000	286500	
Operating Cost	249280	249625	--Inf–
Final Cost	546280	536125	
UB	546280	536125	
Master Solutions			
No of trays	12	11	
Feed tray	6	5	
LB	534330	535890	
UB-LB<Tolerance	NO	NO	STOP

The solution to this problem is summarised in Table 2. The optimal column configuration consists of 12 trays with optimal feed located on 7th tray. The final total cost obtained is 517000 $/yr. The optimal control scheme obtained is pairing reflux flowrate with top temperature of the column (T_c) and steam flowrate with feed tray temperature (T_7). The

noticeable fact, from this result, is that in order to obtain better purity and maximum conversion, in the case of reactive distillation, it is important to control the temperature of the column, both feed tray as well as product (top column temperature, for this particular case). This somehow contrasts with the conventional control scheme in distillation columns, where top and bottom compositions (or temperatures) are controlled in order to attain better performance. Moreover, for this particular case the composition of ethyl acetate is reduced in the bottom of the column by almost 10%, indicating less product losses and better separation when using this control scheme.

Figs. 2 and 3 show the top and bottom product composition at the optimal solution.

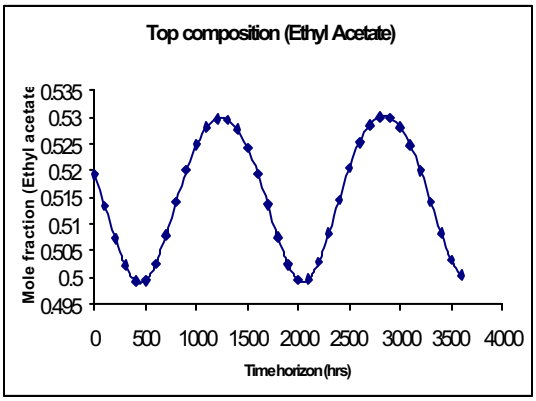

Fig. 2 Ethyl acetate top composition

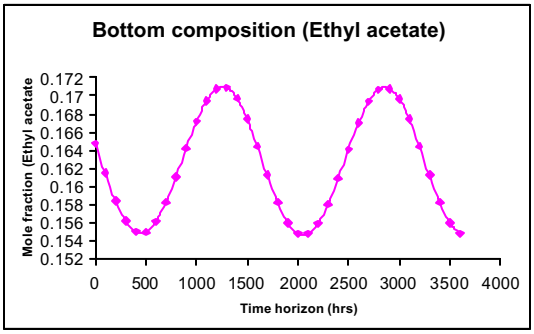

Fig. 3 Ethyl acetate bottom composition

Both figures illustrate that the ethyl acetate composition satisfies the product quality constraints over the entire time horizon (at least 50% of ethyl acetate at the top; and, at the most 20% at the bottom of the column).

The temperature profile of a reactive distillation column is closely related to the extent of reaction or reaction rate. Figure 4 depicts the reaction rate throughout the column on the solution of the MIDO problem. It is observed that most of the reaction is actually taking place at the bottom part of the column. Since reaction takes place mainly in the bottom section of the column with tray 7 (feed tray) having the maximum extent of reaction, a new column configuration could be considered where top trays are considered as non-reactive and bottom trays as reactive. By introducing the binary variables to model reactive and non-reactive trays in the superstructure, it is expected that the performance of the design obtained will be better than that of the fully reactive distillation column.

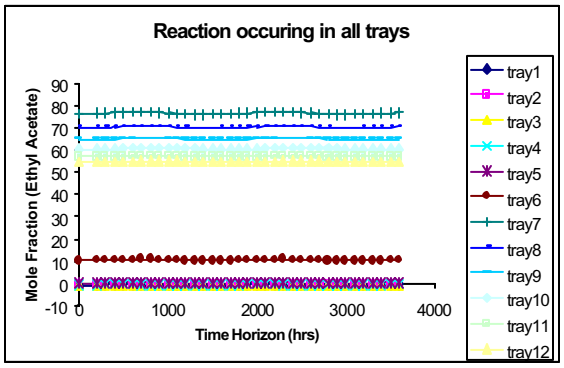

Fig. 4 Reaction profile in all the trays

3.3 Discussion of Results

The results obtained by using integrated MIDO approach are summarised as follows:

- The simultaneous consideration of process design and control results in economic savings of around 20% compared to the original design thus highlighting the importance of addressing the interactions and synergistic benefits that exist between process design and process control in the early design stage. For the case when only process design is considered the savings are around 17%.
- It was also found that by determining the design and control structure simultaneously, savings of almost 3.7% are obtained compared to the case where the control structure is fixed. The reduction in the capital cost and the operating cost are approximately 1.5% and 6% respectively. There is also a considerable reduction in the reboiler duty and the condenser duty, in the order of 10%. The reboiler and the condenser areas are significantly reduced. Moreover, the reflux and steam flow rates are reduced. This strongly portrays the impact that the interactions between design and control have on the operability.
- Simultaneous design and control strategy results in the reduced composition of ethyl acetate in the bottom stream, which implies less product losses, than with fixed control structure strategy.
- For this reactive distillation system, it is required to control the temperature of the reactive tray in which maximum extent of reaction takes place.
- Controlling feed tray temperature emphasizes the fact that the behaviour of reactive distillation column is different than the conventional distillation, where normally the composition of products (or top and bottom temperatures) are controlled to obtain better purity. The temperature in the reactive section needs to be controlled to obtain maximum conversion and better purity of the product.

Table 2 Progress of Iterations for the overall MIDO strategy

Iteration No	1	2	3
Primal Solutions *Discrete Decisions*			
No of trays	13	12	11
Feed location	6	7	3
1st controller	R-$x_{4,d}$	R-T_c	R-$x_{4,d}$
2nd controller	F_s-$x_{4,r}$	F_s-T_7	F_s-T_c
Process Design			
Diameter of column *(m)*	5.00	5.10	
Reboiler surface area *(m2)*	170.00	140.00	–Inf--
Condenser surface area *(m2)*	480.00	470.00	
Controller gains			
1st	25	42.52	–Inf--
2nd	5726.00	5174.00	
Controller Reset times			
1st	60	28.00	–Inf--
2nd	4936.00	4966.00	
Controller Bias			
1st	3065.00	2790.00	
2nd	59000.0	55830.0	–Inf--
Capital Cost	297000	282000	
Operating Cost	249280	235000	–Inf--
Final Cost	546280	517000	
UB	546280	517000	
Master Solutions			
No of trays	12	11	
Feed tray	7	3	
1st controller	R-T_c	R-$x_{4,d}$	
2nd controller	F_s-T_7	F_s-T_c	
LB	445135	470000	
UB-LB<Tolerance	NO	NO	STOP

4. CONCLUSIONS

This work considers an advanced MIDO framework for studying the interaction between process design, process control and operability in a reactive distillation system. The results demonstrate the benefits from pursuing a simultaneous approach in process and control design rather than the traditional sequential approach. Economic advantages of the order of 3.7% are obtained with feasible dynamic performance, production specifications and operational restrictions despite the presence of rapidly varying disturbances.

Although this work has focused on the design of traditional PI controllers, more advanced control schemes (e.g. model-based parametric controllers) could be incorporated based on recent advances in our group (Sakizlis *et al*., 2003; Sakizlis, 2003). It is thus expected that even more inexpensive designs could be achieved with proven dynamic operation and economics over the simultaneous process and control design featuring conventional controllers. Furthermore, the importance of new binary variables in the MIDO framework for the automatic identification of the type of tray (reactive or non-reactive) is expected to lead to more economic benefits since the cost of catalyst would be reduced.

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