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A hybrid optimization method (GA/SQP) for methyl acetate production process by reactive batch distillation

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The initialization problem of classical optimization techniques (for instance, Sequential Quadratic Programming) is generally crucial for the robustness of the code. This paper presents a contribution to handle this kind of problems by implementation of a hybrid optimization technique based on the successive use of a Genetic Algorithm (GA) and of a classical Sequential Quadratic Programming (SQP) method: the GA is used to perform a first search in the solution space and to locate the neighbourhood of the solution. Then, the SQP method is employed to refine the solution. The optimization of methyl acetate production process by reactive batch distillation serves as an illustration to validate the approach.

1. Introduction

Batch processes are widely carried out for the production of fine chemical or pharmaceuticals with generally many successive reaction and separation steps involved. Synthesis optimization is often limited to the determination of the optimal operating conditions of each step separately. Therefore, such an approach does not lead to the optimal conditions for global synthesis. For example, conversion optimization of a reaction for which separation between the desired product and the by-products is more difficult than between the reactants will involve an important operating cost, due to further difficulties in the separation scheme. Thus, the simultaneous integration of all the process steps in a single optimization approach is of major importance (Wajge and Reklaitis, 1999, Elgue et al., 2002).

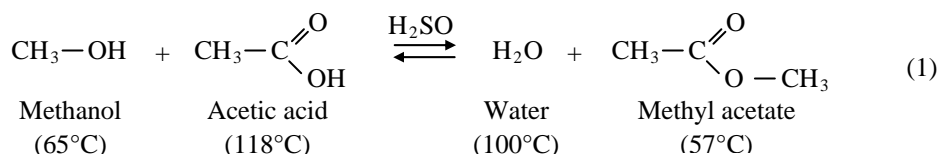
The purpose of the study is to optimize the reactive distillation process of methyl acetate production. A simulation model was previously developed and validated for describing this process (Elgue et al., 2002). The model was further used in connection with an optimization loop involving an objective function combining two criteria, i.e. operating time and conversion rate with various weights.

This study has shown that the initialization problem of classical optimization techniques which may be used (for instance, Sequential Quadratic Programming) is crucial for the robustness of the code. For this purpose, an optimization strategy combining a two-stage approach, i.e. a genetic algorithm for initialization purpose and identification of the search zone followed by an SQP method to refine the solution is proposed in this paper: the GA is used to perform a first search in the solution space and to locate the neighbourhood of the solution. Then, the SQP method is employed to refine the solution. In this paper, the basic principles of the model development will be briefly recalled. The main results of the classical optimization approach by SQP will be presented. Then, the basic operations involved in the design of the GA developed in this study (encoding with binary representation of real values, evaluation function, adaptive plan) are presented. The performances of the method are analyzed and compared with the only SQP procedure.

2. Model presentation

A general model taking into account the various configurations of reaction separation process was developed (Elgue et al., 2002). The mathematical model involves a rigorous description of a batch reactor connected with an overhead distillation column and is formulated as a set of differential and algebraic equations (DAE system) around each plate.

Ordinary differential equations (ODE) are due to energy balances, total and component mass balances. Algebraic equations (AE) consist of vapour-liquid equilibrium relationships, summation equations and physical property estimations. To reduce the complexity of the model, the following typical assumptions have been made on each tray: perfect mixing between vapour bubbles and liquid, equilibrium between liquid and vapour bubbles, introduction of Murphree efficiency, negligible vapour holdup compared to the liquid holdup and constant volume of liquid holdup. The mathematical model is connected to Prophy physical property estimation system (enthalpy model, equilibrium constant, hydrodynamic relationship and bubble point temperature) with associated data bank. DISCo (Sargousse et al., 1999), a general solver of DAE systems based on the Gear method is used to solve the mathematical model. The model is presented in detail in (Elgue et al., 2002) for methyl acetate production by reactive distillation according to the following reaction:



Production of methyl acetate is carried out by addition of acetic acid to methanol with sulfuric acid as homogeneous catalyst, as it can be shown on the reaction scheme (1).

A good agreement was obtained between simulations and experimental runs, thus validating the model assumptions and confirming interest in process optimization.

For this purpose, let us recall that operating time, reaction yield, energy consumption, safety and environmental constraints are generally the key points of reactive batch distillation processes. In the concerned process, the column is always operating with a total reflux policy during reaction phases and with a constant reflux policy during distillation.

Thus, energy consumption (heat provided by the boiler, condenser cooling water) is only a function of the operating time. In this way, process optimization only involves two criteria, i.e. operating time and conversion of reactants. Therefore, a function combining operating time and conversion rate with various weights has been proposed in order to optimize the production process. Hence, a mono-objective optimization method based on a Sequential Quadratic Programming method (SQP) was used.

All the different processes, reaction-distillation or their combination have the same dynamic structure (see Figure 1). In fact, according to the total reflux end time ($t_{R\infty}$), before or after the time of reaction equilibrium (t_{eq}), the process is respectively a coupling or a reaction-distillation process. Thus, the total reflux end time appears to be a main variable of the optimization problems, as well as reflux ratio (R) and operating time (t_{op}).

The total reflux end time is a very sensitive parameter of reactive batch distillation. On the one hand, if total reflux policy ends too early, conversion will be reduced owing to reactant distillation. On the other hand, a too long total reflux policy will lead to

additional operating time and, consequently, to waste of time. An adequate value for total reflux time is very difficult to estimate experimentally and optimization represents a very challenging objective for its automatic determination.

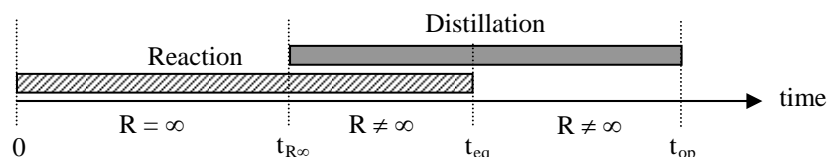


Figure 1: Dynamic structure of batch reactive distillation process

3. Optimization with SQP package

Two kinds of optimization problems were investigated (see Table 1). In the former, the minimum operating time necessary to obtain the desired reactant conversion is determined. In the latter, the objective functions involve a combination of operating time and conversion with various weights.

Table 1: Optimization problems

	Objective function	Variables	Constraints
1.	Operating time	Operating time Total reflux end time Reflux ratio	Model equations Acid Conversion
2.	Operating time, acetic acid conversion	Operating time Total reflux end time Reflux ratio	Model equations

Several optimization problems were solved with SQP method (SQP package from IMSL library) (Schittkowski, 1986). The main results are presented in Table 2. The two first solutions present the optimal operating conditions for respectively 93 percent and 95 percent of acetic acid conversion (C_{onv}). The three other solutions are relative to operating conditions leading to optimal criteria, for which the conversion weight decreases respectively.

Results of the first set of optimization problems show that a significant total reflux time (more than 15 min) is required for a high conversion of reactants. The second set show that if conversion is favoured, total reflux time is almost invariable (around 23 min) and only further operating time allows reaching a higher conversion.

Table 2: Results of the optimization problems (with SQP)

Objective function	Constraint	Operating time (min)	Total reflux end (min)	Reflux ratio	Acetic acid conversion
t_{op}	$C_{onv} \geq 93 \%$	80	15	1.8	93.0 %
t_{op}	$C_{onv} \geq 95 \%$	118	20	3.0	95.0 %
$t_{op} + (1 - C_{onv}) \times 33$	/	141	25	2.3	95.9 %
$t_{op} + (1 - C_{onv}) \times 17$	/	105	23	2.5	94.5 %
$t_{op} + (1 - C_{onv}) \times 1.7$	/	48	9	1.0	85.8 %

4. Optimization with a GA/SQP hybrid optimization method

A typical feature of complex optimization problems is that the generation of starting points that are used in the search with conventional optimization methods (for example SQP) is very important to guarantee the success of the optimization procedure.

The preliminary study has shown that the only use of an SQP method is very sensitive to the choice of the initial guess and often leads to a failure. Consequently, this work is motivated by the development of a technique for the automatic generation of good starting points.

For this purpose, a hybrid optimization method is proposed in this paper: a Genetic Algorithm (GA) is used to perform a first search in the space and to locate the neighborhood of the solution. Then, the derivative driven optimization tool (SQP) is used to refine the solution. Let us recall that Genetic Algorithms (Goldberg, 1989) differ from most classical optimization methods since no assumption about the problem space is required and yet produce a global search. GAs use a guided random search in which many different solutions to a problem are investigated and refined simultaneously to identify near-optimum solutions. A major interest of such methods is that they lead to reasonable solutions even with a poor initial guess. GA implementation requires the definition of parameters: population generation mode, population size, (i.e., the number of individuals forming a population which must be sufficiently large to create sufficient diversity, in order to cover the possible solution space) crossover probability, mutation rate, survival, crossover and mutation mechanisms. In this paper, the initial population has been selected randomly. Nevertheless, constraint respect at a precision of 5.10^{-6} is required. Each potential solution, i.e., a real value, has been coded as an haploid chromosome with the so-called "weight box". The weight box consists to encode each digit of real in 4 bits with respectively the following weights 1, 2, 3, 3 with a precision of 10^{-20} . By lack of place (see Montastruc et al., 2002), the choice of weight is not justified here but has been achieved after a sensitivity analysis. For example, the real 0.18 is encoded 1000 0111, the zero digit and the point are not encoded. Each chromosome represents the 3 unknowns.

Since a traditional GA is based on a function maximization, the evaluation function has been adapted to also consider a minimization case. More precisely, the treated problem involves both operating time minimization and conversion rate maximization. Moreover, as initially proposed by Goldberg (1989) and followed by several authors (Costa and Oliviera, 2001), the penalty function method has been used to take into account the constraints in the optimization procedure. Two cases have thus been taken into account:

$$\begin{aligned} \text{Evaluation function} &= (t_{\max} - t_{\text{op}}) + P X_{\text{conv}}, \text{ for } t_{\text{op}} > t_{R\infty} \\ \text{Evaluation function} &= (t_{\max} - t_{\text{op}}) + P X_{\text{conv}} - P_2 (t_{R\infty} - t_{\text{op}})^2, \text{ for } t_{\text{op}} < t_{R\infty} \end{aligned}$$

In these expressions, $t_{R\infty}$ is the time from which a change in reflux ratio occurs. The constraint $t_{\text{op}} - t_{R\infty} > 0$ is thus taken into account. If this constraint is not satisfied during individual selection phase, another penalty term is introduced (coefficient P_2). Consequently, the individuals will have lower chances to be selected and to contribute to the next generation.

5. Results

5.1 GA parameter setting

The definition domain of the variables which has been taken into account is as follows: $0.6 < r < 0.9$; $1.5 < R < 9$ with $R = r / 1-r$; $0 < t_{op} < 130$ min; $5 \text{ min} < t_{R\infty} < 50$ min
A preliminary study with the GA leads to the following parameter set, which has been arbitrarily fixed, following classical guidelines: Generation number = 40; Population number = 40; Crossover rate = 75%; Mutation rate = 25 %.

5.2 GA+SQP typical results

Four different values of the penalty coefficient P were considered : P=100;1000, 1700 and 2500. By lack of place, only the results relative to P=1700 are presented (See Table 3). They show that the results obtained by SQP are very similar to those obtained by coupling GA and SQP, which shows that the GA is relatively efficient. It has been pointed out that an increase in penalty coefficient P may lead to significant variations in results, which mean that several solutions may exist.

Table 3: Results of GA and GA+SQP for P=1700

Run		Reflux ratio	Operating time	Total reflux time	Conversion rate (%)
1	GA	2,02	120 min	12 min	95,00
	GA+SQP	2,58	90 min	10 min	93,66
2	GA	1,68	120 min	24 min	95,07
	GA+SQP	1,60	120 min	24 min	94,94
3	GA	2,12	109 min	17 min	94,65
	GA+SQP	2,14	107 min	18 min	94,65
4	GA	2,48	106 min	24 min	94,56
	GA+SQP	2,75	107 min	19 min	94,65

Another investigation was performed coupling GA and SQP at different stages of the GA evolution, i.e., for the best individuals obtained respectively at the 4th, 10th, 12th and 20th generations (NG). The results are proposed in Table 4. It can be seen that SQP converges to the same solution for NG=10 and NG=20 whereas initialization is not the same. Yet, different results are obtained for NG=4 and NG=12, which means that the number of potential solutions may be high. It must also be noted that the computational time for GA is significantly higher, which confirms that the use of GA at its early stages of evolution is particularly interesting for determination of initialization values, which are generally difficult to find by trial-and-error procedures.

6. Conclusions

In this paper, a hybrid optimization tool combining a Genetic Algorithm and an SQP method has been developed and tested on optimization of methyl acetate production process by reactive batch distillation. This two-level strategy leads to very efficient search: the GA has been able to provide good starting points for the subsequent SQP method (even if the user has provided rough starting points), thus favouring the local search. The generic feature of the optimization strategy presented in this study finds a widespread application for hybrid optimization problems.

Table 4: Results of GA and GA+SQP for different stages of A evolution (NG, number of generation)

NG=4	Reflux rate	Operating time	Total reflux time	Conversion rate (%)	Evaluation function	Computation time
GA	3,46	126 min	15 min	95,18	- 676 041,5	20 min
GA+SQP	3,72	126 min	15 min	95,21	- 676 254,4	23 min
NG=10						
GA	2,38	100 min	15 min	94,23	- 676 819,0	45 min
GA+SQP	2,68	100 min	15 min	94,34	- 677 650,1	52 min
NG=12						
GA	2,38	91 min	15 min	94,11	- 677 122,6	55 min
GA+SQP	2,52	91 min	15 min	94,15	- 677 364,4	59 min
NG=20						
GA	2,56	100 min	15 min	94,32	- 677 500,1	90 min
GA+SQP	2,68	100 min	15 min	94,34	- 677 650,8	93 min

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