

Heterogeneous Batch Distillation Processes: Real System Optimisation

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Abstract

In this paper, optimisation of batch distillation processes is considered. It deals with real systems with rigorous simulation of the processes through the resolution full MESH differential algebraic equations. A specific software architecture is developed, lying on the ProSim BatchColumn simulator and on both SQP and GA numerical algorithms. The efficiency of the proposed optimisation tool is illustrated by a case study of heterogeneous batch solvent recovery by distillation. Significant economical gains are obtained along with improved process conditions. For such a multiobjective complex problems, GA is preferred to SQP that is able to improve specific GA solutions.

Keywords: Optimisation, Batch Distillation, Heterogeneous Azeotrope

1. Introduction

Solvent recovery is a major issue in the pharmaceutical and specialty chemical industries. In that purpose, batch distillation is a separation process of choice. For azeotropic or close-boiling mixtures, the addition of an entrainer, partially or totally miscible with one of the initial binary mixture components, is viable and its choice is the first key issue of azeotropic batch distillation. A whole set of entrainer selection rules has been published for both homogeneous and heterogeneous azeotropic distillation for the separation of azeotropic binary mixtures or close boiling components (Rodríguez Donis, 2001a and 2001b). These rules also hint at a feasible sequence of batch needed to perform the separation together with the initial feed stream location in the ternary diagram. But the optimisation of the batch sequences is a second key issue and this contribution validates a framework for the optimisation of complex distillation.

2. Problem definition

The goal of batch sequences optimisation is to minimise an overall economical criterion while respecting constraints such as purity, It can be considered as a nonlinear constraint optimisation problem. The classical formulation involves an objective function (f), equality constraints and inequality constraints (g_i and h_j respectively):

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$$\begin{aligned}
& \text{Min } f(x) \\
& g_i(x) = 0 \quad i = 1, \dots, n_g \\
& h_j(x) \leq 0 \quad j = 1, \dots, n_h
\end{aligned} \tag{1}$$

2.1. Objective function

The objective function f is the summation of six cost functions c_i :

Table 1. Economical cost functions taken into account in the objective function

cost function	$f = \sum_1^6 c_i$	object	expression	used variable
c_1		immobilisation	$c_1 = a_1.t + b_1$	t = total separation duration
c_2		energy	$c_2 = a_2.Q$	Q = total required energy
c_3		load	$c_3 = a_3.L$	L = global column load
c_4		entrainer	$c_4 = a_4.E$	E = entrainer amount added initially
c_5		column treatment	$c_5 = a_5.R + b_5$	R = residual column load
c_6		tanks treatments	$c_6 = \sum_{k=1}^{n_T} a_6^k.T_k + b_6^k$	T_k = final load of each of the n_T tanks (including still)

2.2. Constraints

The constraints of the NLP problem are defined with respect to target purity and/or quantity specifications at the end of the distillation process.

Each constraint h_j is expressed as follows:

$$h_j = x_i^k - x_{i,obj}^k \tag{2}$$

where x_i^k and $x_{i,obj}^k$ are the effective and target fraction of component i in tank k .

2.3. Action variables

Action variables are chosen among all the available running specifications of the batch process, that is a collection of successive tasks and the initial load of entrainer (Table 2).

Table 2. Available action variables

Available action variable (* for each task i)	
Entrainer load	Task duration *
Boiling duty *	Reflux ratio of light phase *
Subcooling temperature *	Reflux ratio of heavy phase *

3. Problem resolution

3.1. Overall resolution software architecture

The proposed optimisation methodology lies on a rigorous simulation of the considered batch processes. Most of the variables values required to evaluate the objective function and the constraints are calculated through this process simulation. From a defined column configuration and defined initial settings, a full MESH (Material balance,

Equilibrium, Summation of molar fraction, Heat Balance) set of differential algebraic equation is solved using the ProSim BatchColumn software (ProSim SA, France).

Main results from the batch simulations are mass and composition in each distillate tank and in the boiler, as well as the total heating and cooling duties. The economical optimisation criterion and the constraints values is evaluated from these results. These evaluations stand for the heart of the resolution software architecture, as shown in *Figure 1*.

Optimisation algorithms offer strategies to change the values of the action variables in order to solve the constraint minimisation problem.

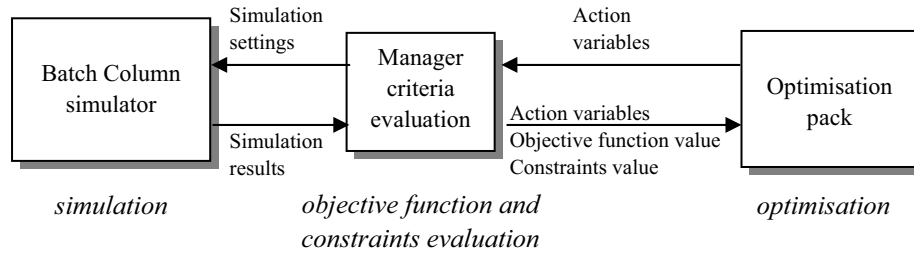


Figure 1. Optimisation software architecture

3.2. Optimisation methods

Two optimisation techniques are used: a SQP-based deterministic method, and a home made Genetic Algorithm as a stochastic one.

The SQP algorithm is the *donlp2* tool, available at www.netlib.org (Spellucci, 1998). It incorporates the exact 11-merit function and a special BFGS quasi-Newton approximation to the Hessian. The optimisation problem is strictly equation (1).

The genetic algorithm is real-coded. In order to use such an unconstrained optimisation technique, the constraints are introduced into the objective function by penalty terms. The optimisation problem aims then at finding the minimum of the following f_p function:

$$f_p = f + \sum P_i (g_i)^2 + \sum Q_i (h_i)^2 \quad (3)$$

$$\text{with } P_i(x) = \begin{cases} 0 & \text{if } g_i(x) = 0 \\ P_i^0 & \text{if } g_i(x) \neq 0 \end{cases} \quad \text{and} \quad Q_i(x) = \begin{cases} 0 & \text{if } h_i(x) \leq 0 \\ Q_i^0 & \text{if } h_i(x) > 0 \end{cases}$$

and P_i^0 and Q_i^0 are weighting factors proportional the inverse of the squared tolerances on the constraints.

4. Separation of pyridine from water using toluene as entrainer

4.1 Problem settings

We study the separation of the minimum temperature homoazeotropic binary mixture water – pyridine. According the Rodriguez-Donis et al. (2002) and Skouras (2004), the separation is possible using a heterogeneous entrainer. Toluene is added to the mixture, forming a minimum temperature homoazeotrope with pyridine and a minimum

temperature heteroazeotrope with water. Three distillation regions exist with the water – toluene heteroazeotrope being the unstable node in each region and the stable node being the pure vertexes. The distillation boundaries are strongly curved and tangent to the vapour line at the heteroazeotrope, like any residue curve in the VLLE region. During the heterogeneous batch rectification process, removal of the aqueous phase in the decanter is expected and reflux of either the non-aqueous phase or a combination of both decanter phases is possible. In this work, the whole non-aqueous decanter phase is refluxed. This operating mode is called Mode B by Skouras (2004) who described elegantly the heterogeneous batch distillation process issue and feasibility in complement to Rodriguez-Donis et al. (2002). According to Skouras (2004), the initial charge composition must be above the line pyridine – aqueous phase to make the process feasible. The batch distillation boundary has no impact on mode B process feasibility (but does on mode A, see Skouras (2004)). The residue curve/distillation boundaries have no impact on feasibility despite their curvature.

Table 3. VLL and LL Thermodynamic parameters (liquid: NRTL; gas: ideal gas)

Parameter value (cal/mol)	A_{ij0}	A_{ji0}	α_{ij0}	A_{ijT}	A_{jiT}	α_{ijT}
Water – Toluene	3809.1	2776.3	0.2	21.182	-7.3179	0
Water – Pyridine	1779.18	416.162	0.6932	0	0	0
Toluene – Pyridine	264.64	-60.34	0.2992	0	0	0

For NRTL: $g_{ij}-g_{ji}=A_{ij0}+A_{ijT} \cdot (T-273.15)$; $g_{ji}-g_{ii}=A_{ji0}+A_{jiT} \cdot (T-273.15)$; $\alpha_{ij}=\alpha_{ij0}+\alpha_{ijT} \cdot (T-273.15)$

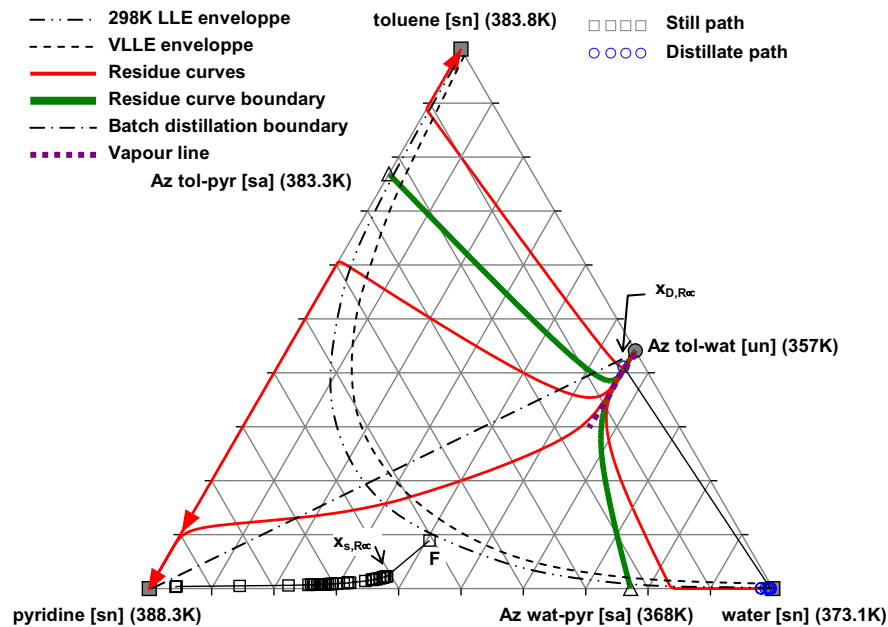


Figure 2. Water/Pyridine/Toluene ternary diagram

The column has 16 plates (incl. decanter). Initial charge of 100 moles with molar fractions [water: 0.4; toluene: 0.1; pyridine: 0.5] is placed in the still. Decanter and plate holdup equals 5 and 1 moles respectively. All plate efficiency are set to unity. Decanter is subcooled to 25°C. Column pressure is 1 atm. and pressure drop is 0.05 atm. The process consists of three tasks: filling, infinite reflux and distillation (water phase removal). Thermodynamic model parameters are given in table 3. These parameters are used to compute VLE in the column and 25°C LLE in the decanter.

The optimisation problem aims at minimising overall costs and satisfying two molar composition inequality constraints:

- $h_1(x_{\text{water}}) > 0.992$ in the distillate tank at the process end.
- $h_2(x_{\text{pyridine}}) > 0.95$ in the still at the process end.

Costs c_1 , c_2 , c_4 , and c_5 (table 1) are used with arbitrary cost factors not detailed here.

Five action variables are defined: entrainer load (F_E); heat duty ($Q_b/R\infty$) and task duration ($t/R\infty$) for the infinite reflux task; heat duty (Q_b/Dist) and task duration (t/Dist) for the distillate removal task.

Tolerances in the constraints are set to 0.001. For the GA, initial population equals 100 to ensure efficient sampling of the five dimensional space; stop criteria equals 0.1. For the SQP, centered gradients are used. Influence of costs factors and optimiser parameters are not considered in this presentation.

4.2 Results and discussion

Results of the optimisation are given in table 4. The initial point does satisfy the constraints on purpose (with respect to tolerances for $h_1(x_{\text{water}})$). If not, results not shown here indicate that the initial criteria is very high because constraint $h_2(x_{\text{pyridine}})$ is not satisfied and its Q_i° factor is proportional to $1/(0.001^2)$. Only the GA finds a solution while the SQP does not.

Table 4. Optimisation results

	initial	SQP	GA	SQP after GA
F_E (kg)	2	1.954	0.866	0.806
$Q_b/R\infty$ (kcal/hr)	3000	2998	1212	1209
$t/R\infty$ (hr)	0.2	0.154	0.043	0.026
Q_b/Dist (kcal/hr)	3000	3000	1437	1433
t/Dist (hr)	0.6	0.533	0.647	0.617
Criteria f	155.468	137.182	82.090	77.882
Criteria f_p	183.453	NA	82.090	NA
$h_1(x_{\text{water}})$	-4.24E-04	-7.10E-05	8.9E-04	1.4E-03
$h_2(x_{\text{pyridine}})$	5.38E-03	9.70E-03	9.1E-03	-5.0E-04
Water purity	0.992	0.992	0.993	0.993
Pyridine purity	0.955	0.960	0.959	0.949
Water recovery	100%	100%	95%	93%
Pyridine recovery	82%	82%	85%	87%
Gain	0%	12%	47%	50%

For results in table 4, the SQP improvement of initial conditions is slight. GA ends after 10 population generations but the after the first generation, the criteria has already

improved to $f=f_p=90.03$ with a mean criteria $f=128.3$ and $f_p=1.0 \cdot 10^6$ indicating a wide sampling of the solution space. An important reduction of the heat duty is observed along with a reduction of the entrainer load. SQP improvement of the GA solution is acceptable.

Of course, results are dependant of the cost factors and problem setting. Indeed, for the separation considered, the entrainer load can be much lower to ensure a feasible separation (see Skouras, 2004). When done with a cost of entrainer c_4 25 times greater than other costs, the optimiser logically finds a best entrainer load equal to 0.294 kg, but heating required is greater. Besides, for a given purity, Q_b/Dist and t/Dist are linked: heating more implies less time to obtain as much water.

On this particular problem and on all problems of batch distillation process optimisation we have performed, the SQP does not perform well when the initial point is infeasible, whereas the GA is always able to find a suitable solution to the problem. In fact, SQP should be used on a feasible solution like the final result of a GA run. This indicates that the GA should always be ran first, unless a specific tuning on a feasible solution is sought.

Simulation of the optimal solution of the SQP optimisation done after the GA (last column) leads to the composition profiles reported in figure 2 for the still and the distillate composition. As can be seen, the distillate at the end of the infinite reflux period lies on the vapour line and rapidly shifts to the LLE envelope at 298K. The still path moves towards the pyridine vertex. Those paths are in agreement with the path already published (Rodríguez-Donis *et al.*, 2002; Skouras, 2004)

5. Conclusion

An optimisation framework coupling stochastic GA and deterministic SQP approaches has been devised and suited for batch distillation processes, homogeneous or heterogeneous, single batch or batch sequence. In the present contribution, validation is done on a double task single rectifier heterogeneous batch process for the separation of water- pyridine with toluene. The five action variable problem is solved. The use of GA followed by an SQP is the recommended choice.

As careful weighting of the action variables shows, such a powerful optimisation tool should be used by users well acquainted with the process expected behaviour.

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Acknowledgements

This work was performed in collaboration with ProSim SA (Toulouse, France), and supported by the french environment and energy agency (ADEME).