CHEMICAL ENGINEERING TRANSACTIONS
VOL. 61, 2017
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.aidic.it/cet
Guest Editors: Petar S Varbanov, Rongxin Su, Hon Loong Lam, Xia Liu, Jiří JKlemeš

ISBN978-88-95608-51-8; **ISSN** 2283-9216
DOI: 10.3303/CET1761211

Global Optimization of Reactive Distillation Processes Using Bat Algorithm

Jiawei Lu[a], Jihai Tang[a,b,c], Xian Chen[a], Mifen Cui[a], Zhaoyang Fei[a,b], Zhuxiu Zhang[a], Xu Qiao[a,b,c,*]

[a]College of Chemical Engineering, Nanjing Tech University, China
[b]State Key Laboratory of Materials-Oriented Chemical Engineering, Nanjing Tech University, China
[c]Jiangsu National Synergetic Innovation Center for Advanced Materials (SICAM), China
qct@njtech.edu.cn

Reactive distillation (RD) is an important process intensification approach with several advantages. It can improve the reaction selectivity and yield, overcome the thermodynamic restrictions, and reduce the cost/energy. However, the optimal design of RD relies on highly nonlinear and multivariable optimization including continuous and integer design variables. The objective function is generally non-convex with several constraints. For this problem, the conventional derivative-based optimization algorithms are faced with convergence problems and fail to guarantee the global optimal solution.
Stochastic optimization algorithms appear to be a better alternative for the optimal design of RD because of the high robustness and efficiency. Bat algorithm (BA), which combines advantages of other existing algorithms, is a potential stochastic optimization algorithm. In this work, the BA was used to optimize the RD for the production of methyl acetate (MeAc). The link between Matlab and Aspen Plus was also created to ensure that each solution was provided from rigorous simulations. The total annual cost (TAC) was set as the objective function. Product purity constraints were achieved through Aspen plus instead of algorithms to simplify the process. BA can find the global optimal solution within less computation time than other stochastic algorithms or sequential optimization.

1. Introduction

Process intensification (PI) is an effect strategy to save resources and energy. The purpose of the PI is to decrease the back mixing, reduce the mass and heat transfer resistances and accelerate the reaction rate in chemical process. (Moulijn et al., 2008) One of the most common process intensification approaches is the reactive distillation (RD), which combines chemical reaction and distillation separation in a device, where the reaction process and the distillation process promote each other. Because of this, it can effectively improve the reaction selectivity and yield, overcome the thermodynamic restrictions and increase the reaction heat efficiency. However, the optimal design of RD is a more complex and difficult problem, which is different from the traditional distillation process.
Ciric and Gu (1994) were first to present a rigorous model based on the reaction kinetic rate equations and the MESH. The optimization problem of RD was transformed into a MINLP problem, which was solved with Generalized Benders Decomposition (GBD). But this formulation exhibited poor numerical behaviour. Zondervan et al. (2011) have developed a MINLP model to optimize the design of RD. The results indicated that DICOPT could find a solution near to global optimality. Harwardt et al. (2011) argued that the MINLP problem of the RD process is challenging and reformulated the MINLP problem as a continuous NLP problem to achieve better convergence. In recent years, several deterministic algorithms are used for the optimization of RD. However, the objective function is generally non-convex with several constraints. Therefore, the optimization process is always faced with convergence problems, and easy to fall into the local optimum.
Stochastic optimization algorithms appear to be a better alternative for the optimal design of RD because of the high robustness and efficiency. A new simulated annealing-based algorithm, which combined the simplex

Please cite this article as: Lu J., Tang J., Chen X., Cui M., Fei Z., Zhang Z., Qiao X., 2017, Global optimization of reactive distillation processes using bat algorithm, Chemical Engineering Transactions, 61, 1279-1284 DOI:10.3303/CET1761211

method of Nelder and Mead, was applied to the MINLP problem. (Cardoso et al., 2000) But this method still cannot guarantee the global optimum. Cheng et al. (2009) studied the simulated annealing for the optimization of RD. This method could find a better design than the sequential design approach and save more computation time. Domingues et al. (2014) studied the particle swarm optimization and genetic algorithm for the ETBE synthesis reaction distillation optimization. The results show that these two algorithms can get the similar structure of the column. Although the stochastic algorithms have the ability to escape the local optimum, it often need to cost great amount computation time. Therefore, how to improve the efficiency of the stochastic algorithm or find a better algorithm has become the focus of the research. (Segovia-Hernández et al., 2015)

This work studied a new stochastic algorithm, bat algorithm (BA). Compared to other existing stochastic algorithm, BA can automatically switch from global search to local search when the conditions are met to reduce the search time. Because of this, the BA, which was implemented in the Matlab, was applied to optimize the reactive distillation for the production of methyl acetate (MeAc). The process simulator, Aspen Plus, was also used for the simulation of RD to ensure the reliability of the calculation.

2. Bat algorithm

The bat algorithm is a new nature-inspired stochastic optimization algorithm, which is based on the echolocation behaviour of bats. (Saji et al., 2016) The points in the search space are considered as the bats in nature. The whole process can be simplified as that the bats fly with velocity v_i, varying frequency f_i and loudness A_i at position x_i to search for prey. They can automatically adjust the frequency and the rate of pulse emission r_i according to the distance between the target and their own.

2.1 Velocity and position vectors of virtual bats

In simulations, the rules have to be defined how the positions x_i and velocities v_i are updated. Because of the particularity of the BA, it has local search and global search in the search process. In a d-dimensional search space, for the global search part, the new positions x_i^t and velocities v_i^t at time step t are given by:

$$f_i = f_{min} + (f_{max} - f_{min})\beta \tag{1}$$

$$v_i^t = v_i^{t-1} + (X_i^t - X^*)f_i \tag{2}$$

$$X_i^t = X_i^{t-1} + v_i^t \tag{3}$$

Where $\beta \in [0, 1]$ is a random vector drawn from a uniform distribution. Here x^* is the current global optimal position, which is located after all the bat positions are sorted. In the simulation, the values of the f_{min} and f_{max} depend on the size of the search space. Initially, the frequency of the bat is achieved by randomization.

For the local search part, once the optimal solution is selected from the current solution, a new local solution for each bat is generated around the optimal solution by random walk:

$$X_{new} = X_{old} + \varepsilon A^t \tag{4}$$

Where $\varepsilon \in [-1, 1]$ is a random number, and A^t is the average loudness of all the virtual bats at time step t. It can be seen that the update of the position of the BA has some similarity to the formula in the standard particle swarm optimization (PSO) and harmony search (HS). The reason is that PSO and HS are the special cases of BA after appropriate simplifications.

2.2 Variations of loudness and pulse emission

Furthermore, the loudness A_i and the rate of pulse emission r_i have to be updated as the iterations proceed. As the iteration increases, the loudness of the virtual bats will gradually decrease, and finally reach the minimum while the rate of pulse emission will increase.

$$A_i^{t+1} = \alpha A_i^t \tag{5}$$

$$r_i^{t+1} = r_i^0 [1 - \exp(-\gamma t)] \tag{6}$$

Where α and γ are constants. For any $0 < \alpha < 1$, $\gamma > 0$, we have

$$A_i^t \to 0, \quad r_i^t \to r_i^0, \quad \text{as } t \to \infty \tag{7}$$

In generally, the choice of parameters requires some experiments. But in our simulations, for simplicity, the values of α and γ are chosen in the range of [0.80, 95]. Initially, the loudness and the rate of pulse emission of each virtual bat are achieved by randomization. For example, the initial loudness A_i^0 can typically be [1, 2], while the rate of pulse emission r_i^0 can be any value in the range of [0, 1].

2.3 Iteration process

The Pseudo-code of bat algorithm is described in following steps:

1. Parameter initialization: population size N, loudness attenuation coefficient α, pulse emission rate increasing coefficient γ, Maximum loudness A_i^0, Maximum pulse emission rate r_i^0 and the maximum number of iteration iterMax.
2. Initialize the position x_i and velocity v_i, calculate fitness, find the current optimal bat and its location x^*.
3. Adjust the frequency to generate a new solution and change the velocity and position, calculate its fitness.
4. If rand > r_i, then walk randomly around the best solution and generate a local solution.
5. If rand < A_i and f (x_{new}) <f (x_i), then accept this new solution and adjust the loudness A_i and the pulse emission rate r_i according to the formula.
6. Arrange the whole bats to find the best bat individual and its location
7. If the termination condition is met, output the optimal bat and the global optimal solution. Otherwise, go back to step 3 to proceed the next search.

3. Case studies

This work studied the optimal design of RD for the production of methyl acetate (MeAc), which has been studied in previous work. Tang et al. (2005) used the sequential optimization approach and Cheng et al. (2009) used the simulated annealing algorithm. Both of them obtained the satisfied results.

3.1 Reaction kinetics and phase equilibria

The esterification of the acetic acid (HAC) with methanol (MeOH) is expressed in the following form:

$$MeOH + HAc \rightarrow MeAc + H_2O \tag{8}$$

The reaction kinetics are shown below (Tang et al., 2005):

$$r_c = m_{cat}(k_1 \alpha_{HAc} \alpha_{MeOH} - k_{-1} \alpha_{MeAc} \alpha_{H_2O}) \tag{9}$$

$$k_1 = 2.961 \times 10^4 \exp(-49190/(R \times T)) \tag{10}$$

$$k_{-1} = 1.348 \times 10^6 \exp(-69230/(R \times T)) \tag{11}$$

Where r_c represents the reaction rate; m_{cat} represents the mass of the catalyst; k_1 and k_{-1} represent the reaction rate constants, [kmol/(kg*s)]; α_i represents the activity of component i.

MeOH-HAc-MeAc-H2O quaternary system exhibits a nonideal phase behavior. To predict the nonideal vapor-liquid equilibrium (VLE) and possible vapor-liquid-liquid equilibrium (VLLE) behaviors in this quaternary system, the UNIQUAC model is used to calculate the activity coefficients. Furthermore, Hayden-O' Conell second virial coefficient model is also adopted to describe the dimerization of acetic acid.

3.2 Design variable

In the methyl acetate system, both acetic acid and methanol are assumed as single feed. In the process, there are several variables that need to be determined, including column pressure, acetic acid feed stage (NF_{HAc}), methanol feed stage (NF_{MeOH}), reactants feed ratio (FR), total number of the stage (N_{stage}), reaction section beginning stage (NR_{start}), reaction section ending stage (NR_{end}), the liquid holdup on reactive stage (H), catalyst loading (M_{cat}), reflux ratio (RR), reboiler duty (QR),as shown in Figure 1. The optimization is based on the following assumptions:

1. The RD column is operated under atmospheric pressure and the pressure drop is ignored.
2. The feed flow of methanol and acetic acid are 50 kmol/h.
3. Methanol and acetic acid are fed at the bubble point.
4. The catalyst loading volume is 50 % of the tray holdup volume.
5. The catalyst density is 770 kg/m^3.
6. The purity of the products are 98 mol % (X_{MeAc}=98 mol % and X_{H2O}=98 mol %)

To simplify the whole process, product purity constraints are achieved through Aspen Plus by adjusting the reflux ratio and reboiler duty. The remaining variables of the RD column need to be optimized. Univariate analysis is used to determine the range of values for these six variables, as shown in Table 1.

Table 1: Range of variables

Variable	N_{stage}	NR_{start}	NR_{end}	H (m^3)	NF_{HAc}	NF_{MeOH}
Optimization range	25~45	1~ N_{stage} -2	NR_{start} +1~N_{stage} -1	0.01~0.2	1~ N_{stage}	1~ N_{stage}

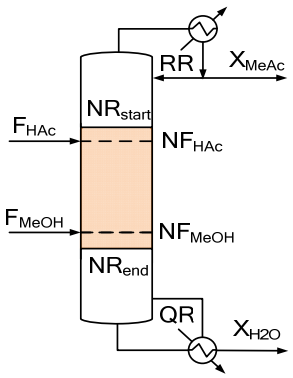

Figure1: Process flowsheet and the design variables for the MeAc System

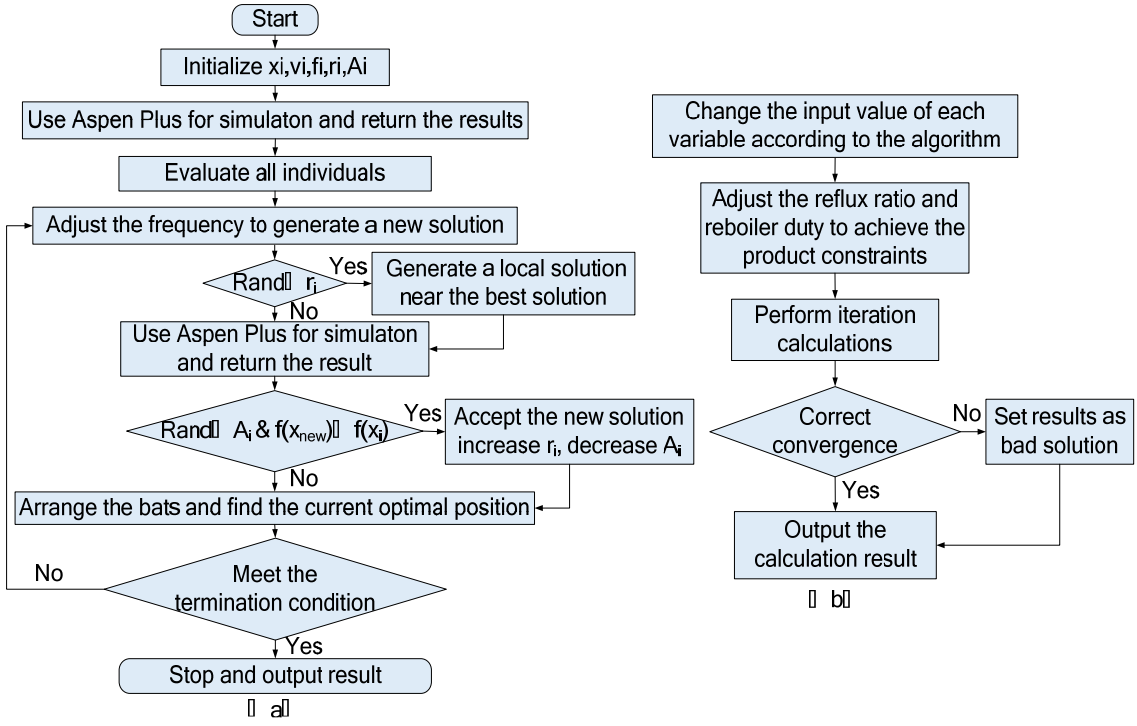

Figure 2: (a) Flowchart of the BA algorithm; (b) Flowchart of the Aspen Plus

3.3 Process optimization

The total annual cost（TAC）is set as the objective function based on Douglas' book, which includes capital cost and operation cost.

$$TAC = \text{Capital cost}/\text{Payback period} + \text{Operation cost} \tag{12}$$

Where the capital cost is the combination of the column and heat exchanger. And the operation cost covers the cost of steam, cooling water and catalyst. In this study, the payback period is set as 3 y and the cost of piping and pumps are ignored.

In the simulation, the BA was implemented in the Matlab, which could transfer data with Aspen Plus by COM technology. During the iterative process, to ensure the stability of the algorithm, the points that were not converged in the Aspen Plus would be set as bad solution and removed from the iteration.

3.4 Results and discussion

Before optimization，the parameters of the BA have to be determined. The bat population was set as 150 after several tests. As the population increases, the results would be better. But it would cost more computation time. There are several constant parameters of the bat algorithm, including maximum frequency f_{max}, minimum frequency f_{min}, loudness attenuation coefficient α, pulse emission rate increasing coefficient γ, initial loudness A_0, maximum pulse emission rate r_0. Both A_0 and r_0 were generated randomly in their respective ranges. Generally, the f_{min} is set as 0 and the f_{max} is determined by the size of the solution space. When f_{max} is set as a large value, it is benefit to enhance the global search ability, but the bat will rock around the optimal solution at the end of search. On the contrary, when f_{max} is set as a small value, it is helpful to search accurately, but it will fall into local optimum easily. Additionally, α and γ are always in the range of [0.80, 95]. The effect of these parameters for a better optimization was explored in this work.
First, the f_{max} was fixed of 4 to study the effect of α and γ by using three different cases (cases A, B, C), and the results are given in Table 2. The results show that α and γ have no significant effect on the optimization process. The results of three different cases are similar and the deviation is less than 2 %. The stability of Case B is better than case A and C. Because of this, the α and γ was set as 0.9.

Table 2: Optimization results with different parameters

Case	N	iterMax	α	γ	Run	TAC (10^3\$)	Dev.
A	150	50	0.85	0.95	1	236.8	1.50 %
					2	235.7	1.03 %
B	150	50	0.90	0.90	1	233.3	0 %
					2	233.7	0.17 %
C	150	50	0.95	0.85	1	234.1	0.34 %
					2	237.5	1.80 %

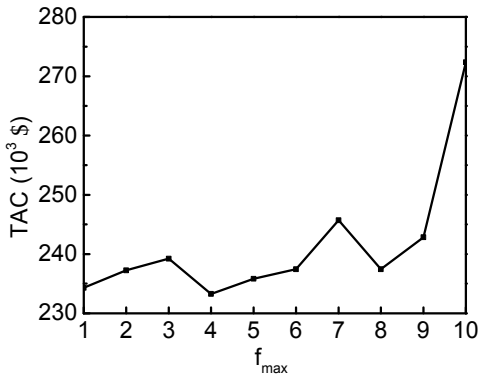

Figure 3: The optimal solution for the MeAc system with different maximum frequency f_{max}

The maximum frequency f_{max} controls the velocities of the bats, and the effect of it was studied by setting it from 1 to 10. As shown in the Figure 3, the results that we get are satisfied until the value is greater than 6. And when it is set as 4, the best solution could be obtained. So, the f_{max} was set as 4.
Figure 4 (a) shows that the reliable design can be achieved by using bat algorithm. As the number of iteration increases, the objective function (TAC) appears to decrease significantly at the beginning and it becomes stable in the end. These three runs do not converge to the same value because of the high nonlinearity of the system, but the deviation of them is not large. All of them can be used as a reliable design.

Table 3: Results of the MeAc system by bat algorithm

Variable	N_{stage}	NR_{start}	NR_{end}	H(m^3)	NF_{HAc}	NF_{MeOH}	TAC(10^3\$)
Results	25	17	22	0.08	5	20	233.3

The results were also compared with two other optimization methods, as shown in Figure 4 (b). One design was optimized by simulated annealing (Cheng et al., 2009), and the other one was obtained by sequential optimization approach (Tang et al., 2005). However, the former method ignored the effect of the liquid holdup, and the later method required a good initial condition. The results show that the BA can achieve the solution

better than these two methods. (Evaluation function is same.) Authors cannot guarantee that this is the global optimal solution, but it is also a satisfied design. Authors think that when the solution can meet the design requirement and have a high quality, it can be used as a global optimal solution to save computation time.

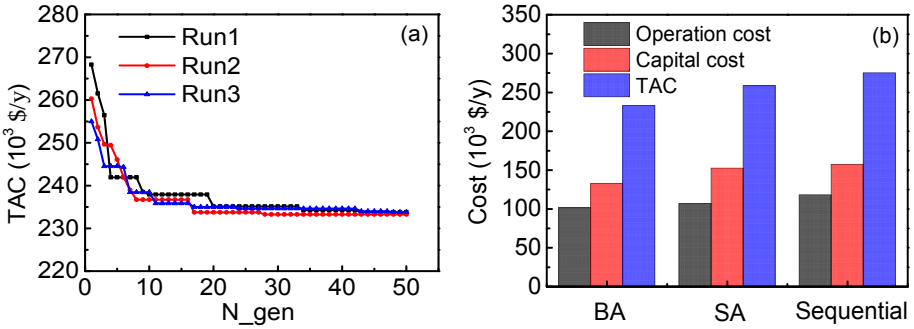

Figure 4: (a) Results of bat algorithm process for the MeAc system; (b) Comparisons of different optimization methods for the MeAc system

4. Conclusions

In this work, the BA was applied to optimize the reactive distillation for methyl acetate. The results indicate that BA is feasible in optimization and it can improve the quality of the solution. When it is coupled with process simulators, the reliability of the solution can be guaranteed. In addition, the BA, which is potentially more powerful than other methods, can be further improved. Authors made a simple exploration about the effect of the parameters of the BA, and it can become the basis for the bat algorithm improvement in the future. Because of the superiority of the BA, authors believe that it can be applied to more chemical processes.

Acknowledgments

This work was supported by the Natural Science Foundation of China (21276126, 21676141 and 61673205), National key R&D Program of China (2017YFB0307300), Six Major Talent Peak Project of Jiangsu Province (XCL-017), The Fifth Period Project "333" of Jiangsu Province (BRA2016418) and Prospective Joint Research Project of Jiangsu Province (BY2015005-02)

References

Cardoso M.F., Salcedo R.L., Azevedo S.F.D., Barbosa D., 2000, Optimization of reactive distillation processes with simulated annealing, Chemical Engineering Science, 55(21), 5059-5078.

Cheng J.K., Lee H.Y., Huang H.P., 2009, Optimal steady-state design of reactive distillation processes using simulated annealing, Journal of the Taiwan Institute of Chemical Engineers, 40(2), 188-196.

Ciric A.R., Gu D., 1994, Synthesis of nonequilibrium reactive distillation processes by MINLP optimization, AIChE Journal, 40(9), 1479-1487.

Domingues L., Oliveira N.M.C., 2014, Optimal design of reactive distillation systems: Application to the production of ethyl tert-butyl ether (ETBE), Computers & Chemical Engineering, 64(7), 81-94.

Harwardt A., Kraemer K., Rüngeler B., Marquardt W., 2011, Conceptual design of a butyl-levulinate reactive distillation process by incremental refinement, Chinese Journal of Chemical Engineering, 19(3), 371-379.

Moulijn J.A., Stankiewicz A., Grievink J., Górak A., 2008, Process intensification and process systems engineering: A friendly symbiosis, Computers & Chemical Engineering, 32(1-2), 3-11.

Saji Y., Riffi M.E., 2016, A novel discrete bat algorithm for solving the travelling salesman problem, Neural Computing and Applications, 27, 1853-1866.

Segovia-Hernández J.G., Hernández S., Petriciolet A.B., 2015, Reactive distillation: a review of optimal design using deterministic and stochastic techniques, Chemical Engineering & Processing Process Intensification, 97, 134-143.

Tang Y.T., Chen Y.W., Huang H.P., Yu C.C., Hung S.B., Lee M.J., 2005, Design of Reactive Distillations for Acetic Acid Esterification, AIChE Journal, 51(6), 1683-1699.

Zondervan E., Shah M., 2011, Optimal design of a reactive distillation column, Chemical Engineering Transactions, 24, 295-300.