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Optimal Design of a Reactive Distillation Column

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In this work we develop a MINLP model that can be used to optimize the design of reactive distillation column. MINLP model is formulated in GAMS in such a way that it can be solved locally and globally. In the RD column a component A is converted into product B while vapour and liquid are assumed to be in equilibrium. The objective is to find a design for this process that minimizes the total costs (consisting of capital and operational costs). The design variables of interest are the total number of stages, the number of reactive stages, the location of the reactive stages, the feed tray location and the reflux ratio.

Keywords: Reactive distillation, Design, Optimization, MINLP

1. Introduction

Reactive distillation (RD) is a matured technology that combines reaction and separation in a single processing unit. RD has distinct advantages; normally the equipment is much smaller than conventional equipment, the energy requirements are lower and the conversion of the product is higher as the products are immediately removed by distillation. *Krishna et al.* (2002) give a more complete overview of reactive distillation.

However, design and control of RD is a complex process (*Al-Arfaj and Luyben* (2000)). Especially the optimal design of such a system requires accurate process models that lead to a computationally demanding mathematical problem.

Although the problem has been studied in the scientific literature, most of the time the proposed models are strong simplifications of reality and most authors agree that the more complex models cannot be solved to global optimality

In *Jackson and Grossmann (2001)* an optimization approach for the optimal design of a reactive distillation column is proposed, which shows that disjunctive programming can be effectively used to handle the resulting nonlinear optimization problem. The design of a reactive distillation column is concerned with finding the total number of trays of the column, the number and location of reactive trays, and the feed and reflux locations of the column.

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Also Seferlis and Grievink (2001) solve a similar problem using collocation models. Stochastic optimization methods such as genetic algorithm are also often applied to design these processes. However, this approach is computationally expensive and because of a probabilistic approach, there is no assurance of global optimality.

This model is associated with nonlinearities from reaction kinetics, phase equilibrium and bilinear terms of the balance equations. *Gangadwala et al.* (2006) have formulated MINLP model for RD process. However, they can only solve the problem locally. For global optimality they have applied polyhedral relaxations and converted MINLP problem to MILP problem. They have concluded that MINLP problem for RD process can only be solved locally.

To overcome this design problem, in this work an RD model is formulated as MINLP in such way that model can be solved globally. This model contains continuous- as well as discrete variables. Continuous variables are usually related to operating conditions such as liquid and vapour flows, feed flows, reflux ratio. Discrete variables are related to number and positions of reactive stages, reflux location, number and positions of feed, required stages to obtain pure product.

2. Problem statement and proposed model

In this section an MINLP is proposed to optimize the design of a reactive distillation column. The optimization objective is to minimize the total costs and to find optimal reboiler and condenser duties, the reflux ratio, the number of stages, the number and location of reactive stages, catalyst loadings on reactive stages and the feed location. In the column a reaction A ↔ B takes place for which the reaction kinetics, component balances and material balances are known, also vapour and liquid are assumed to be in equilibrium for the system of our interest.

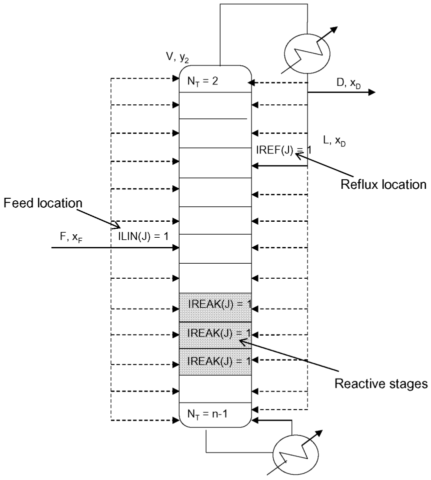

Figure 1: A schematic view of RD column and graphical view of discrete binary variables

A schematic view of RD column is shown in figure 1 which also includes all important design variables to be determined by solving the MINLP model. The stages are numbered from top to bottom. The first stage represents condenser and the last stage represents the reboiler. Since there is only one product produced in a column, which is obtained as distillate, a total condenser is used to obtain the distillate at the top of a column. A reactant is heavy component and unreacted reactant has to be recycled back completely to the column thus a total reboiler is used, which results RD column without bottom flow rate.

The binary variables such as *IREAK (J)* for reactive stage location, *IREF (J)* for reflux location, *ILIN(J)* for feed location are introduced to know whether a stage *J* is a reactive stage *(IREAK (J) =1)* or a top stage receiving reflux *(IREF (J) =1)* or feed stage *(ILIN (J) =1)*. Liquid is not present on the stages above the reflux stage so these stages have no effect on the column performance. Hence, the total number of stages is calculated as: $N = N_{\max} - \sum J \cdot IREF(J) + 2.0$. The summation of binary variable *IREAK (J)* gives total number of reactive stages.

The objective function which represents the total cost of reactive distillation column is based on the column dimensions and the heat duties:

$$\min f(N_T, H, D, \Delta T) \tag{1}$$

Where N_T is the total number of stages, H is the column length, D is the column diameter and ΔT are the heat duties. The component balances at each stage n can be given as:

$$g_n(L, V, x, y, r) = 0, \forall n \tag{2}$$

Where L are the liquid flows, V the vapour flows and x and y the liquid and vapour compositions. The reaction kinetics holds that:

$$r_n = h_n(x), \forall n \tag{3}$$

And for the vapour-liquid equilibrium we use a relative volatility relation of the form:

$$y = k_n(x) \tag{4}$$

The model also includes logical constraints to incorporate only one feed and one reflux stage:

$$\sum(n_F) = 1 \qquad \sum(n_{RR}) = 1 \tag{5}$$

and the constrains for the reflux stage above the feed stage:

$$\sum J \cdot ILIN(J) > \sum J \cdot IREF(J) \tag{6}$$

Furthermore the model includes structural constraints that ensure the operational conditions, e.g. flows cannot exceed certain minimum and maximum values, or the configuration settings such as the number of reactive stages cannot exceed the total number of stages. To ensure that the product at the outlet has a specified purity we introduce

$$x_D \geq x_p \tag{7}$$

where x_P is the requested product purity. Eqs. 1-7 above form a mixed integer nonlinear programming problem (MINLP) and nonlinearities are associated with reaction kinetics, phase equilibrium and bilinear terms of the balance equations and product purity.

3. Results and discussions

A pure component *A* is fed to the column and a minimum product purity of 99.5% of component *B* in distillate is set as a constraint. The simulation of the reactive distillation model is performed with the characteristic system data given in table 1.

Table 1: Modelling data

Parameters	**Description**	**value**
F	feed flow rate (Kmol/hr)	1.5
Kf0	forward reaction rate constant [Kmol/kg/hr]	3.33e04
Ef0	activation energy for forward reaction [KJ/kmol]	57.69e03
kb0	backward reaction rate constant [Kmol/kg/hr]	2.24
Eb0	activation energy for backward reaction [KJ/kmol]	8.47e03
T	temperature of column [K]	335
P	pressure [bar]	1.013e02
MW	molecular weight [Kg/kmol]	84.16
HETP	height equivalent to theoretical plate	0.33
ΔTREB	temperature gradient for heat transfer at condenser [K]	35.0
ΔTCON	temperature gradient for heat transfer at reboiler [K]	35.0

Since the product is obtained as distillate, it can be seen from figure 2 that the composition of the product is high at the top stage compared to composition of reactant. The composition of reactant is high at the bottom stage because reactant is heavy component and recycled back to bottom of the column. The optimal design variables are tabulated in table 2. The optimal design encompasses a reflux ratio of 6.32, and a total of 29 stages are required to produce 99.5% pure product at the top of the column. The optimal design suggests introducing a feed to the column at 28th stage. In total 18 reactive stages are required and these reactive stages are located at stage 12 to 29 in the column. The total costs of this system are 1.41e05 USD to produce 800 tons per year. In particular, 1.10e05 USD is the capital cost of a reactive distillation column and 3.06e04 USD is the operating cost of the column.

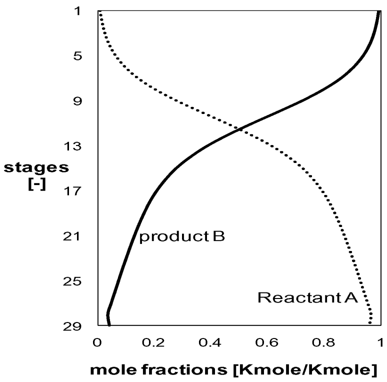

Figure 2: liquid compositions profile of reactant and product along the column

Table 2: optimal design variables found from simulation

Variables	Descriptions	value
D	diameter (m)	0.489
H	height of column (m)	12.91
A_{reb}	required area for reboiler (m^2)	3.185
A_{cond}	required area for condenser (m^2)	2.954
Q_{reb}	required heat for reboiler (KJ/hr)	3.21e05
Q_{cond}	required heat for condenser (KJ/hr)	2.98e05
M_{CAT}	required catalyst loadings per reactive stage (kg)	9.284
RR	reflux ratio (kmol/kmol)	6.32
LR	reflux rate (kmol/hr)	9.382
V	vapour flow rate (kmol/hr)	10.867
L	liquid flow rate (kmol/hr)	9.382

The MINLP formulation of RD model contains 260 equations, 253 continuous variables and 87 binary variables. This resulting MINLP problem is solved using standard optimization tools in GAMS. For local optimization, particularly DICOPT is used with MINOS for the NLP sub problems and CPLEX for the MIP sub problems. To evaluate whether DICOPT has found the global optimum, the MINLP model is ran with a global optimization solver called BARON. The local optimization solvers requires upper and lower bounds for variables but the global optimization solver does not require bounds for variables, which indicates that the solution obtained in this case is at its global optimum. We found the optimal design of RD column with DICOPT in 0.28 seconds and only 28 major iterations are required. BARON found the same design as DICOPT and solved the problem to global optimality in 4673 seconds (5361 iterations). BARON requires more iterations compared to DICOPT because variables are not bounded for BARON and thus BARON tries to check all possible combinations in order to ensure the global optimality. The computational results of two different solvers are compared in table 3.

Table 3: Solver comparison for MINLP problem of reactive distillation column

	DICOPT	**BARON**
Objective value	1.41e+05	1.41e05
CPU time (sec)	0.281	4673
Total number of iteration	28 (major iterations)	5361
Best solution found at node	-	3583

4. Conclusions

We have developed a MINLP model for the optimal design of a reactive distillation column. Numerical results are presented and the formulated problem is subsequently solved with DICOPT and BARON. DICOPT performs considerably faster than BARON, while the found objective values are identical; indicating that DICOPT can finds a solution near to global optimality.

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