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Reactive distillation: A review of optimal design using deterministic and stochastic techniques

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ABSTRACT

In the last years, the industries have shown interest in the development of reactive separation processes. Reactive distillation can be considered as reaction and distillation combined into one new unit operation and this integrated separation process is a good example of process intensification. This intensified process offers several important advantages that include the increment of the reaction yield and selectivity, the overcoming of thermodynamic restrictions and the considerable reduction in energy, water and solvent consumptions. Therefore, this process configuration has been applied in several chemical industries. However, due to the strong interactions of chemical reactions and heat and mass transfer, the design of this intensified separation process tends to be quite complex. The design of reactive distillation systems can be performed using single and multi-objective optimization approaches. This paper provides a comprehensive short review on current applications of deterministic and stochastic optimization techniques for the design of reactive distillation. Capabilities and limitations of optimization for reactive distillation design are discussed and topics for future research are provided.

Contents

1. Introduction

Process intensification (PI) is an effective strategy to achieve increased energy efficiency. PI aims at reducing the mass and heat transfer resistances while overcoming thermodynamic limitations through the integrated design and operation. In recent years, PI has attracted considerable academic interest as a potential means for process improvement, to meet the increasing demands for sustainable production [44,52,49]. A variety of intensified operations developed in the academia and industry creates a large number of options to potentially improve the process performance. However, the identification of the set of feasible solutions for PI in which the optimal condition can be found, may take considerable resources and it can be considered as a challenging task [48,10].

One of the most common examples of the process intensification field is the reactive distillation (RD), where the integration of reaction and separation is performed (Fig. 1). In all the cases where reactive distillation has been used, the variable cost, capital expenditure and energy requirement are reduced by 20% or more, when compared to the classic set-up of a reactor followed by

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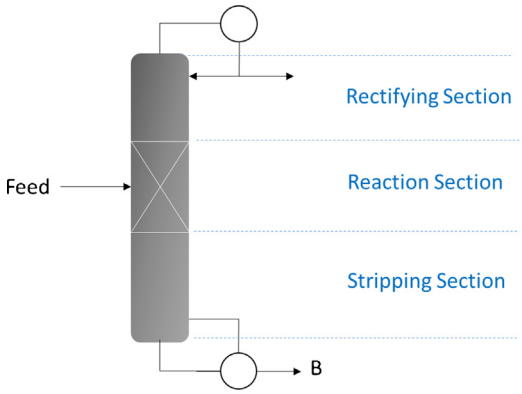

Fig. 1. Reactive distillation column.

distillation [27,64,59]. The first patents of RD, which were for the application of homogeneously catalyzed esterification, are of 1920s [5,6]. The first heterogeneously catalyzed process was patented for the production of methyl tert-butyl ether [61]. Although invented in 1921, the industrial application of reactive distillation did not take place before the 1980s [39]. Classic success stories in reactive distillation are the Eastman Chemical Co.'s methyl acetate reactive distillation process and the processes for the synthesis of fuel ethers. Some of the obtained improvements are so dramatic, for example, five times lower investment and five times lower energy use for the Eastman process [60]. The commercial success of reactive distillation for the production of MTBE was immediately followed by other remarkable achievements [63] and the last few years have seen a dramatic rise in the number of applications of RD. This useful technology is now being applied for any scale of operation from manufacture of fine chemicals to that of bulk chemicals. Applications for RD include esterification, transesterification, hydrolysis, etherification, hydrogenation, dehydrogenation, alkylation, metathesis and disproportionation, hydration and dehydration, carbonylation, production of polymers, alcoxysilanes and acyloxysilanes production, chlorination and amination, synthesis of carbonates, application for chiral separation, recovery of chemicals, and miscellaneous reactions such as those involved in the production of phenol, propylene oxide process, synthesis of vinyl acetate, among others [36].

Basically, RD is a process where the chemical reactor is also the separator. The concept of combining these two important functions for the enhancement of process performance is considered an important contribution in the chemical engineering community. Separation of the product from the reaction mixture does not need a separate distillation step, which saves energy (for heating) and materials [63]. This separation process is particularly attractive and useful for equilibrium-limited reactions. Therefore, it can be suitable for the disproportionate reactions because it eliminates conversion and phase equilibrium limitations. Note that conversion can be increased far beyond what is expected by the equilibrium due to the continuous removal of reaction products from the reactive zone [63]. This helps to reduce the capital and investment costs and this process may be important for sustainable development due to a lower consumption of resources [41]. It is convenient to remark that the suitability of RD for a particular reaction depends on various factors such as volatilities of reactants and products along with the feasible reaction and distillation temperature. Hence, the use of RD for every reaction may not be feasible [19–21]. Therefore, the exploring of the candidate reactions for RD is an area that needs considerable attention to expand the domain of RD processes for other promising industrial applications.

Being a relatively new field, the research on various aspects of RD design such as modeling and simulation, process synthesis, column hardware design, non-linear dynamics and control is in progress. In particular, the design of RD columns focuses on the identification of the characteristics and operating conditions of the separation system to improve its performance, cost, profitability, safety, reliability and other attributes of interest. The design issues for reactive distillation systems are significantly more complex than those involved in ordinary distillation. For example, the catalyst selection, liquid holdup on each tray, and position of feeds become important design considerations. Reaction often occurs in the liquid holdup so that the reaction volume is a major design parameter, and constant molar overflow cannot be assumed. Also, a single feed may not be appropriate and a distributed feed must be considered. The design complexity increases with the configuration of the separation system (e.g., Petlyuk or thermally coupled columns) due to the increment on the degree of freedoms.

Results reported in the literature showed that the optimization strategies can be reliably used for facing this relevant design problem in chemical engineering. In particular, the optimization methods are important tools for process modeling, synthesis, design, operation and retrofitting of separation systems (Rangaiah and Bonilla-Petriciolet [74]). However, the design of RD columns is a complex optimization problem with challenging features for current optimization strategies. Overall, the design of RD systems involves several degrees of freedom with both continuous and discontinuous design variables and the presence of non-linear and potentially non-convex objective functions (i.e., characteristics or performance criteria to be maximized or minimized), which can be subjected to both equality and inequality constraints. Therefore, the research in the application of improved optimization methods for RD design has grown significantly during last years.

This review analyzes and describes the application of optimization techniques for the design of reactive distillation columns. This work provides a survey of different deterministic and stochastic optimization methods applied for solving this design problem. Capabilities and limitations of current numerical strategies have been discussed, including single and multi-objective optimization methods, and topics for further research are also described.

1.1. Formulation of the design of reactive distillation columns as an optimization problem

The objective of this section is to define the general formulation of the optimization problem for the design of RD columns. Overall, this design problem can be stated as follows

$$\text{Optimize}\left\{f_1(\vec{x}), f_2(\vec{x}), \ldots, f_{n_{\text{obj}}}(\vec{x})\right\} \tag{1}$$

subject to

$$g_i(\vec{x}) \leq 0 i = 1, 2, \ldots, n_{\text{ine}} \tag{2}$$

$$h_i(\vec{x}) = 0 i = 1, 2, \ldots, n_e \tag{3}$$

$$\vec{x}_L < \vec{x} < \vec{x}_U \tag{4}$$

where f_i is the objective function i (i.e. attribute or characteristics) to be optimized (i.e., maximized or minimized) during the RD column design, n_{obj} is the number of objective functions involved in the RD design, $\vec{x}$ is the vector of m decision variables (continuous and/or discontinuous) with lower ($\vec{x}_L$) and upper ($\vec{x}_U$) bounds, n_{ine} and n_e are the number of inequality (g) and equality (h) constraints, respectively. The feasible space for solving the design optimization