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Systematic Design of Chemical Reactors with Multiple Stages via Multi-Objective Optimization Approach

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Abstract

The purpose of this study is to extend the approach that was introduced by Hillestad (2010) to handle chemical reactor design problem with multiple stages. Specifically, multi-objective optimization method will be used to generate Pareto optimal solutions that characterize the non-inferior solutions set for the problem. Following the identification of path-dependent design variables, several (possibly conflicting) design objectives will be selected and solutions of the corresponding problem will be generated from multi-objective optimization algorithm. This approach is investigated for two industrially important reactor systems: ethylene oxide and phthalic anhydride synthesis. By using reference-point based multi-objective evolutionary algorithm (R-NSGA-II), Pareto-optimal solutions are successfully generated within the region of user-specified reference points, thus facilitating in the selection of final optimal designs. Apart from the extensive selection of optimal candidate reactor designs, this approach also enables further insights to be obtained regarding the optimal arrangement of the path-dependent design variables along the reactor length.

Keywords: systematic staging; chemical reactor design; multi-objective optimization; R-NSGA-II; ethylene oxide; phthalic anhydride.

1. Introduction

Chemical reactors have always been considered as the heart of chemical processes. Improved reactor design and operation will usually translate into significant cost savings and revenue potentials for the chemical plants. Various analytical methods and design strategies have been introduced to handle the complexity nature of these problems. Among such methods are attainable region, phenomena vectors, and superstructure optimization. Recently, Hillestad, (2010) has proposed a chemical reactor design that is based on a systematic staging concept along the reactor path. In this concept, the reactor path is divided into stages where each stage is designed so as to optimize a single objective function. Additional path-dependent design functions can be introduced for this purpose, for example, fluid mixing profile, distribution of extra feed points, coolant temperature profile, catalyst dilution, and many more. In this approach, the design problem is converted into an optimal control problem where the optimal path designs become the additional degree-of-freedom for optimization. This approach has the potential to derive novel reactor design that can improve further the performance of the process.

There are many trade-offs that must be considered carefully when designing a chemical reactor. Among these trade-offs are balancing the reactor size with reactant conversion, balancing reactant conversion with selectivity, or balancing heat transfer area with hot spot temperature. Suitable reactor design parameters must be selected so that these trade-offs can be balanced in an appropriate way. The multi-objective nature of this problem can be handled systematically if multi-objective optimization algorithm is adopted. In this approach, these trade-offs will become the objective functions to be optimized simultaneously while the algorithm will be used to generate non-inferior solutions along the Pareto-optimal front of these objective functions. The application of this approach for reactor design problem has been investigated for several important industrial reactor systems in the past. Some of these examples are styrene reactor system (Yee et al., 2003), steam reformer (Rajesh et al., 2000), and ethylene reactor (Tarafder et al., 2005).

The purpose of this study is to apply multi-objective optimization approach to derive optimal reactor design that employs systematic staging along the reactor path. Two industrial reactor systems are adopted to illustrate this method, i.e. ethylene oxide and phthalic anhydride synthesis. The corresponding reactors resemble a multi-tubular fixed-bed reactor with constant coolant/wall temperature. It will be shown that systematic staging concept that is coupled with multi-objective optimization algorithm will enable derivation of novel reactor design that can improve further the performance of the process in various aspects.

2. Mathematical modelling and optimization formulation

2.1. Plug-flow model

All the reactors in this study are treated as a plug-flow system to simplify the mathematical representation for optimization purpose. As the optimization step that is adopted in this work is a time-consuming process, the choice of this simpler representation will be justified front the point of view of minimizing the complexity of mathematical model solution. If required, the results can be further validated by simulating the reactor system with more complicated mathematical model later. However, the effort is being made also to validate the approach by comparing the obtained solutions with industrial reactor design that is simulated by the same mathematical model that is used for optimization. The following mathematical models are adopted to represent the corresponding reactor systems in this study:

Ethylene oxide synthesis reactor (Peschel et al., 2011):

$$\frac{dn_i}{dz} = L_t A_c \alpha \rho_b \sum_{j=1}^{N_R} \nu_{i,j} r_j \tag{1}$$

$$\frac{dT}{dz} = L_t A_c \frac{-\alpha \rho_b \sum_{j=1}^{N_R} r_j (-\Delta H_j) - \frac{4}{d_i} U (T - T_{cool})}{\sum_{i=1}^{N_I} n_i C_{pi}} \tag{2}$$

$$\frac{dP}{dz} = -L_t \left[150 \frac{\mu u_0 (1-\varepsilon)^2}{d_p^2 \varepsilon^3} + 1.75 \frac{\rho_f u_0^2 (1-\varepsilon)}{d_p \varepsilon^3} \right] 10^{-5} \tag{3}$$

with initial conditions $z = 0 \rightarrow n_i = n_{i0}$, $T = T_0$, and $P = P_0$.

Phthalic anhydride synthesis reactor (Orozco et al., 2010):

$$u_s \frac{dP_a}{dz} + L_t \left(\frac{\rho_b M_m P_t}{\rho_g}\right) r_a = 0 \quad (4)$$

$$u_s \rho_g C_p \frac{dT}{dz} - L_t \left[(-\Delta H)\rho_b r_a + \frac{4U}{d_t}(T - T_w)\right] = 0 \quad (5)$$

with initial conditions $z = 0 \rightarrow P_a = P_{a0}$ and $T = T_0$.

The kinetics expressions for the corresponding reactions are taken from the respective literatures: ethylene oxide synthesis (Al-Saleh et al., 1988) and phthalic anhydride synthesis (Anastasov, 2003).

2.2. Path-dependent design variables

A path is a line along the reactor where the reactions take place (Hillestad, 2010). Along this path, additional design variables can be introduced that act as the forcing function to modify the characteristics of the reaction process taking place. The path-dependent design variables for the respective reactor systems are summarized in Table 1. The corresponding variables are parameterized by piecewise constant profiles. This method divides the reactor path into several stages, N_z where the length and design variable for each stage is optimized.

Table 1. Path-dependent design variables for the respective reactor systems

Reactor system	Path-dependent variables	Number of stages
Ethylene oxide synthesis	Catalyst dilution	2
Phthalic anhydride synthesis	Catalyst types – low productive catalyst (Catalyst I) versus high productive catalysts (Catalyst II and Catalyst III)	3

2.3. Objective functions

The multiple objective criteria that are selected for each reactor designs are summarized in Table 2. These criteria are selected based on the profitability, safety, and cost minimization aspect of the reactor design. It can be seen that these criteria are conflicting with each other, thus requiring necessary trade-off in the final reactor design.

Table 2. Design objectives for the respective reactor systems

Reactor system	Design objectives	Target
Ethylene oxide synthesis	Ethylene oxide productivity	Maximize
	Heat transfer area	Minimize
	Active catalyst mass	Minimize
Phthalic anhydride synthesis	Phthalic anhydride composition	Maximize
	Mass of high productive catalysts	Minimize
	Total reactor volume	Minimize

2.4. Multi-objective optimization formulation

The multi-objective optimization problem for the reactor designs can be represented generally by the following mathematical statement:

$$optimize\left(f_1(x), f_2(x), \ldots, f_n(x)\right) \quad \text{objective functions vector} \quad (6)$$

s.t.

$$g_i(x) \leq 0 \quad i = 1, 2, \ldots, n_i \quad \text{inequality constraints} \quad (7)$$

$$h_i(x) = 0 \quad i = 1, 2, \ldots, n_i \quad \text{equality constraints} \quad (8)$$

$$x_l \leq x \leq x_u \quad \text{decision variables bound} \quad (9)$$

where n is the number of objective functions to be optimized simultaneously, x is the vector of m decision variables (continuous and/or discontinuous) with lower (x_l) and upper (x_u) bounds, n_i and n_e are the number of inequality (g) and equality (h) constraints, respectively. The lists of decision variables and constraints for the respective reactor systems are presented in Table 3.

Apart from the path-dependent design variables, the following reactor design parameters (i.e. tube length, tube diameter, and number of tubes) are also included as additional decision variables for the optimization. Tube diameters and number of tubes are specified by discrete variables so as to follow precisely the allowable range of variation for the corresponding reactor design parameters. The inequality constraint for the minimum exit pressure is also specified to ensure that allowable pressure drop is maintained throughout the reactor length.

Table 3. Decision variables and constraints for the reactor design optimization

Reactor system	Decision variables	Constraints
Ethylene oxide synthesis	α_k, L_k	$0.5 \leq \alpha_k \leq 1$ $\sum_k^{N_z} L_k = L_t$ $T_{max} < 553.15$
Phthalic anhydride synthesis	γ_k, L_k	$\gamma_k \in \{Cat_I, Cat_{II}, Cat_{III}\}$ $\sum_k^{N_z} L_k = L_t$ $T_{max} < 703.15$

2.5. Solution by using R-NSGA-II

As opposed to single-objective optimization that only gives single optimal solution, solutions of multi-objective optimization problems give rise to Pareto-optimal solutions that characterize the non-inferior solutions set for these problems. These solutions have the property such that when one moves from one solution to another in the corresponding set, one objective function improves while the other worsens. In this work, reference- point based multi-objective evolutionary algorithm method (R-NSGA-II) (Deb et al., 2006), a modification of the well-known and popular multi-objective genetic algorithm method (i.e. NSGA-II) is adopted to solve the corresponding reactor design problems. The modification of the original NSGA-II algorithm allows the generation of Pareto-optimal solutions near the region of user-specified reference points, thus facilitating the selection of final optimal solutions. More information regarding the algorithm implementations can be found from their original literatures (Deb et al., 2002; Deb et al., 2006).

3. Results and discussion

Pareto-optimal solutions obtained from R-NSGA-II runs are shown in Fig. 1 for each reactor system considered. The corresponding Pareto solutions are obtained after 200 generations with population size 50, crossover probability 0.7, and mutation rate 0.3. The ε parameter that controls the spread of solutions near the chosen reference points for R-NSGA-II strategy is set at 0.01. The corresponding reference points and the representative industrial reactor designs are also shown in the same figures for comparison purpose. These Pareto solutions offer extensive selections of candidate reactor designs that can be found within the region of user-specified reference points. These solutions also offer insights into the optimal arrangement of the path-dependent

design variables along the reactor length. For ethylene oxide reactor design case, the analysis of the selected Pareto solution clearly indicates that the location of catalyst dilution zone should be near the reactor inlet (see Table 4). The benefits of this configuration are that total heat transfer area can be reduced without increasing the hot spot temperature beyond acceptable limit while at the same time the productivity of ethylene oxide can be further increased. For phthalic anhydride reactor design case, the examination of the selected Pareto solution clearly indicates that the optimum staging of catalyst types can improve further the phthalic anhydride (PA) exit composition if compared with the industrial reactor designs where only one type of catalyst is employed throughout the reactor (see Table 5).

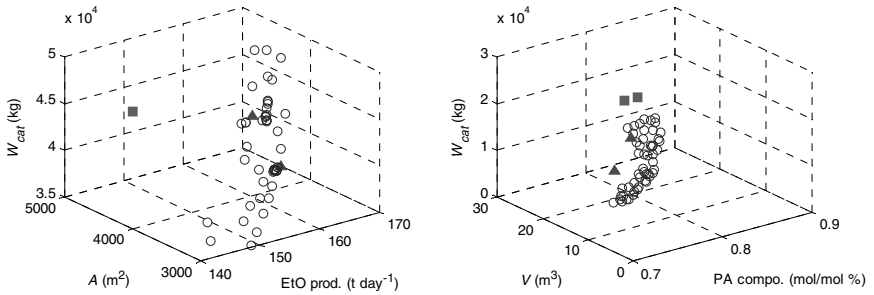

Fig. 1. Pareto optimal solutions for multi-objective optimization of ethylene oxide and phthalic anhydride reactor designs. Pareto solutions (circles), reference points (triangles), and industrial reactors (squares)

Table 4. Comparison of selected Pareto solution with industrial reactor (ethylene oxide reactor design case)

Reactor parameters	Industrial	2-stage configuration
Design parameters:		
Tube length (m)	8	6.19
Tube diameter (mm)	$d_o = 38.1$, $d_i = 31.3$	$d_o = 44.5$, $d_i = 42.7$
Number of tubes	5160	4068
Catalyst per reactor (kg)	42484	41823
Total heat transfer area (m^2)	4941	3519
Path-dependent design variables:		
α_k		$\alpha_1 = 0.5$, $\alpha_2 = 1$
L_k (m)		$L_1 = 1.64$, $L_2 = 4.55$
Operating parameters:		
Ethylene productivity (t d^{-1})	150.87	159.49
Hot spot temperature (K)	555.69	552.97

4. Conclusion

Systematic staging of chemical reactor design via multi-objective optimization approach has been investigated in this study. By demonstrating the application on two industrially important reactor systems (i.e. ethylene oxide and phthalic anhydride synthesis), it can be shown that optimal reactor designs can be obtained that utilized optimally the path-dependent design variables along the reactor length. These results show promising improvement to the reactor design and operation. Moreover, the analysis of the multi-objective optimization results also offers valuable insights into the optimal arrangement of the selected path-dependent design variables that can improve further the

performance of the reaction system. This multi-objective optimization approach is applicable to design other industrial fixed-bed reactors operated under non-isothermal condition.

Table 5. Comparison of selected Pareto solution with industrial reactor (phthalic anhydride reactor design case)

Reactor parameters	Industrial			3-stage configuration
Design parameters:				
Tube length (m)		2.8		2.84
Tube inner diameter (mm)		26		35.5
Number of tubes		10550		5579
Catalyst per reactor (kg)		23525		23486 (Cat. I = 45.2 %, Cat. II = 4.46 %, Cat. III = 50.34 %)
Total heat transfer area (m^2)		2654		1893
Total reactor volume (m^3)		15.68		15.66
Path-dependent design variables:				
γ_k				γ_1 = Cat. III, γ_2 = Cat. I, γ_3 = Cat. II
L_k (m)				$L_1 = 1.43$, $L_2 = 1.28$, $L_3 = 0.13$
Operating parameters:	Cat. I	Cat. II	Cat. III	
PA composition (mol mol^{-1} %)	0.733	0.785	0.771	0.81
Hot spot temperature (K)	727.10	703.19	688.09	701.43

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