Synthesis of Optimal Serial Reactor Structures for Homogeneous Reactions

Part I: Isothermal Reactors

The reactor synthesis problem can simply be stated as follows: Given the reaction mechanism and kinetics, what is the optimum type and arrangement of the reactor(s) that would maximize product yield? Extensive studies for simple reaction systems (series, parallel, consecutive, etc.) using idealized plug flow and continuous stirred tank reactors have been presented in the literature. However, these studies fail to provide a systematic procedure for selecting the reactor(s) type and the arrangement for complex reaction schemes.

In this paper simple rules with geometrical interpretations have been developed for synthesizing isothermal reactor configurations for complex reaction schemes. A computer program capable of specifying the optimum reactor structure from a generalized configuration has also been described.

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SCOPE

What is the optimum type(s), arrangement, and size of the reactor(s) that would maximize product yield for a given reaction mechanism? This is the essence of the reactor synthesis problem in the chemical process industry.

Previous studies in the reaction engineering field have resulted in some guidelines for simple reactions. These rules and their limitations have been presented in this paper. However, these studies fail to provide a systematic procedure for selecting the reactor(s) type and the arrangement for complex reaction schemes.

This paper is the first in a two-part series which attempts to classify reaction schemes into three basic types. Simple rules with geometrical interpretations have been presented for Type II reactions and a synthesis computer program has been described for Type III reactions. These rules are limited to isothermal reactor systems only. Non-isothermal studies will be presented in Part II of this paper.

CONCLUSIONS AND SIGNIFICANCE

In this paper reaction schemes have been classified into three basic types based on the kinetic equations: Type I (simple reactions), and Types II and III (complex reactions).

Simple rules for Type I reactions have been presented in this paper. These rules are based on previous studies by Denbigh (1961), Carberry (1966), and others. Rules have been derived for several serial reactor configurations for Type II reactions. Geometrical interpretations for these rules have been presented to provide an understanding of the mathematics involved. Finally, for Type III reactions, a computer program has been discussed which is capable of specifying the reactor type(s), arrangement, and size for a given reaction mechanism.

This work is significant in that it is the first attempt to develop rules for synthesizing reactor configurations for complex reaction schemes. It provides a methodology for specifying the optimum structure of the reactor system for complex reactions which would be necessary in the synthesis and development of chemical process flowsheets.

INTRODUCTION

The design of chemical process reactors has been recognized for a long time as an important and difficult problem, because invariably the reactor is the most crucial unit operation. Even when the temperatures, catalyst, and basic kinetics are already fixed by the chemistry of the process, the reactor configuration can have a significant effect on the conversion efficiency of many reactions and on the quantity and quality of recycle, which in turn affects process and energy costs.

The design of chemical process reactors can be viewed as a three

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stage iterative process: (1) the selection or modification of the reactor system structure (selection of reactor types and arrangement to maximize product yield and minimize reactor cost) and the identification of the design variables in this structure; (2) the assignment of numerical values to these design variables; (3) evaluation of the resulting design, and optimization, resulting in decisions regarding which of steps (1) and (2), if either, must be repeated.

The design procedure is difficult because chemical reactions with several by-products are inherently complex; because the reaction rate is a nonlinear function of temperature, pressure, and composition variables and hence is affected by heat and mass transfer rates; and because evaluation of the design depends on several objectives including safety, product yield, stability, economics, and ease of start-up, operation, and shut-down.

Traditionally, the selection of the reactor type has been based on the basic idealized reactors, namely the ideal plug flow reactor (PFR) and the ideal continuous stirred tank reactor (CSTR). Most industrial reactors either conform to these basic two types or can be represented by a combination of idealized PFRs and CSTRs. Extensive studies for simple reaction systems (series, parallel, consecutive, etc.) using idealized plug flow and continuous stirred tank reactors have been presented in the literature (Levenspiel, 1962). Studies on representing industrial reactors by a series-parallel combination of idealized PFRs and CSTRs also have been extensively presented in the literature (Keairns and Manning, 1969; Himmelblau and Bischoff, 1968; Olson and Stout, 1966).

However, these studies fail to provide a systematic procedure for selecting the reactor(s) type and arrangement for complex reaction schemes. This is the essence of the reactor synthesis problem that is addressed in this paper. The reactor synthesis problem can simply be stated as follows: Given the reaction mechanism and kinetics for homogeneous reactions, what is the optimum type and arrangement of continuous reactor(s) that would maximize product yield?

In this paper the objective function used to obtain the optimum reactor structure is maximization of the final product concentration (yield). It has been shown by Kondelik (1982) that only for a very inexpensive raw material, a very expensive reactor, and a very low production rate will the reactor cost become significant in comparison to the raw material cost. In a vast majority of cases, the operating costs of a reactor are negligible compared to the raw material cost. Hence the optimum reactor structure depends mainly on the raw material conversion rather than on reactor costs.

BACKGROUND

A detailed review of the literature reveals that extensive studies for simple homogeneous reactions (irreversible first order, second order; reversible; etc.) and complex homogeneous reactions (consecutive reversible and irreversible, parallel reactions, Van de Vusse reaction, etc.) in specified reactor types such as plug flow, continuous stirred tank reactor, and recycle reactor, exist in the literature. These studies are presented briefly in Table 1.

There is an extensive body of papers in the reaction engineering literature that should be referred to in connection with this paper. While this is no place for a general review, we have attempted to present as succinctly as possible previous work that has a direct impact on our work. In addition, to place our work in perspective we have summarized the relevant background in Table 2.

TABLE 1. A BRIEF REVIEW OF THE RELEVANT LITERATURE

Author(s)	Comment
Levenspiel (1962) Van de Vusse (1964)	Reported heuristic rules on yield and selectivity in stirred tank and tubular reactions.
Gillespie & Carberry (1966)	Considered an intermediate level of mixing for the Van de Vusse reaction and showed that in some cases a recycle reactor provides optimum product yield compared to a PFR or CSTR.
Lee (1977)	Demonstrated by a specific example that product yield is higher if the recycle stream originates from an intermediate point along the reactor length.
Chitra & Govind (1981)	Presented optimum product yield for the Van de Vusse reaction in a recycle reactor where the recycle stream can originate from an intermediate point along the reactor length.
Lo & Cholette (1977)	Presented results for reactor combination models showing variation of conversion or residence time as a function of mixing level in the system.
Aris (1961)	Optimized combination of reactions for simple reactions using dynamic programming. Presented analytical and graphical condition for optimality of a CSTR + PFR combination.
Trambouze & Piret (1959)	Proposed graphical and analytical criteria for selection of a CSTR or PFR.
Horn & Tsai (1967) Jackson (1968) Ravimohan (1971)	Optimized chemical reactor networks with respect to flow configurations.
Fan (1966)	Optimized isothermal reactor structures using variational techniques.
Hartmann (1979)	Used the structural parameter method for synthesizing optimally structured reactor systems. This integrated reactor structure is essentially the same as the structure used by Umeda and Ichikawa (1972).
Chitra & Govind (1982)	Obtained optimum reactor structure for a complex reaction scheme using a wide range of kinetic constants.

Representation of Reaction Kinetics

Based on the kinetic equations, complex reactions can be classified into three basic types, as shown in Table 3.

Rules for Synthesizing Isothermal Reactor Configurations

A number of qualitative generalizations have been developed by Denbigh (1961), Carberry (1966), and others. Most of these generalizations were derived from studying simple reactions and are useful in specifying single reactor systems. Combinations of these rules, also summarized below, seem to work for some complex reactions. However, in general, the following rules do not apply for complex reactions (Types II and III).

1. Backmixing (as in a CSTR) is beneficial for reactions where the rate increases with conversion (as in autocatalysis and in the case of negative order kinetics).
2. In a consecutive isothermal reaction (e.g., $A \rightarrow B \rightarrow C$), mixing adversely affects the yield of the intermediate B but favors the formation of the ultimate product C.
3. For isothermal simultaneous reactions (e.g., $A \begin{smallmatrix} \nearrow B \\ \searrow C \end{smallmatrix}$) mixing favors the reaction of lowest order.

These rules, in general, do not work for complex reactions. As stated earlier, developing rules for complex reactions has been the subject of our research. For complex Type II reactions we have derived analytical conditions for several typical reactor configurations. These conditions, some with geometrical interpretations are presented in the following sections.

RULES FOR TYPE II REACTIONS AND A SERIAL COMBINATION OF TWO REACTORS

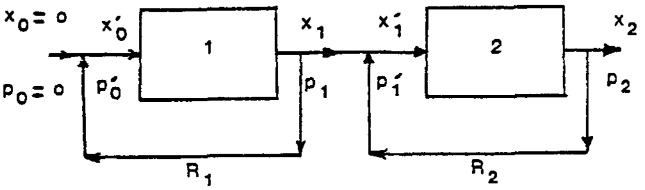
Consider a serial combination of two recycle reactors as shown below, in which the recycle ratio (ratio of volumetric recycle stream flow rate to the volumetric feed rate) of the first reactor is R_1 and that for the second reactor is R_2.

TABLE 2. LITERATURE CLASSIFICATION RELEVANT TO THE SYNTHESIS OF ISOTHERMAL REACTOR SYSTEMS

Mode of Operation	Reactor Type	Simple Homogeneous Reactions	Complex Homogeneous Reactions
	Single Reactor		
	a. PFR	Levenspiel (1962)	Van de Vusse (1964)
	b. CSTR	Rase (1977)	Gillespie & Carberry (1966)
		Paynter & Haskins (1970)	DeVera & Varma (1976)
Continuous Reactors	c. Recycle	Delbridge & Dyson (1973)	Lee (1977)
		Wirges & Shah (1976)	
	Known Combination of Reactors		
	a. *n*-CSTR's in series	Aris (1969)	
		Lo and Cholette (1977)	
		Szepe & Levenspiel (1964)	Aris (1969)
		Wood & Stevens (1964)	
		Burghardt & Bartlemus (1971)	
	b. CSTR + PFR	Aris (1969)	
	c. Mixed Tubular/ Tubular Mixed	Trambouze & Piret (1959)	Chitra & Govind (1981)
			Chitra & Govind (1982)
	Optimum Combination of Reactors	Umeda & Ichikawa (1972)	
		Jackson (1968)	
		Ravimohan (1971)	Chitra & Govind (1982)
		Hartmann (1979)	
		Horn and Tsai (1967)	
	Single Reactor	Fan (1966)	Riddlehoover & Seagrave (1973)
		Codell & Engel (1971)	
		Lund & Seagrave (1971)	Lund & Seagrave (1971)
		Roth et al. (1979)	
		Wagmare & Lim (1981)	
Semi-Batch Reactors	*Cascade of Reactors*	Hussain & Kamath (1968)	

TABLE 3. CLASSIFICATION OF CHEMICAL REACTIONS

Type	Kinetic Equation	Comment
I (Simple Reaction)	$\frac{dp}{dt} = f_1(r,r_0)$ (1)	Product rate is a function of the reactant concentration only.
II (Complex Reaction)	$\frac{dp}{dt} = f_2(p,r,p_0,r_0)$ (2)	Product and limiting reactant rate is function of the product and limiting reactant concentrations only.
	$\frac{dr}{dt} = f_3(p,r,p_0,r_0)$ (3)	
III (Complex Reaction)	$\frac{dp}{dt} = f_4(p,r,\bar{s},p_0,r_0,\bar{s}_0)$ (4)	Product and limiting reactant rate depends on the concentrations of all species.
	$\frac{dr}{dt} = f_5(p,r,\bar{s},p_0,r_0,\bar{s}_0)$ (5)	



The dimensionless concentrations of the product and reactant are designated p and r, respectively, and were defined earlier in the section on reaction kinetics representation (Table 3).

The exit product concentration p_2 can be written as

$$p_2 = p_1 + (R_2 + 1)\int_{x_1' = 1 - r_1}^{x_2 = 1 - r_2} \phi_R(p,x)dx \tag{6}$$

where $\phi_R = dp/dx$ instantaneous yield for a recycle reactor, and

$$x_1' = (x_1 + R_2x_2)/(R_2 + 1) \tag{7}$$

For maximum exit product concentration, p_2, the necessary conditions are

$$\frac{\partial p_2}{\partial x_2} = \frac{\partial p_2}{\partial x_1} = 0 \tag{8}$$

$$\frac{\partial p_2}{\partial R_2} = \frac{\partial p_2}{\partial R_1} = 0 \tag{9}$$

Differentiating Eq. 6 using the Leibnitz rule and substituting Eq. 8, the following necessary condition is obtained for Type II reactions:

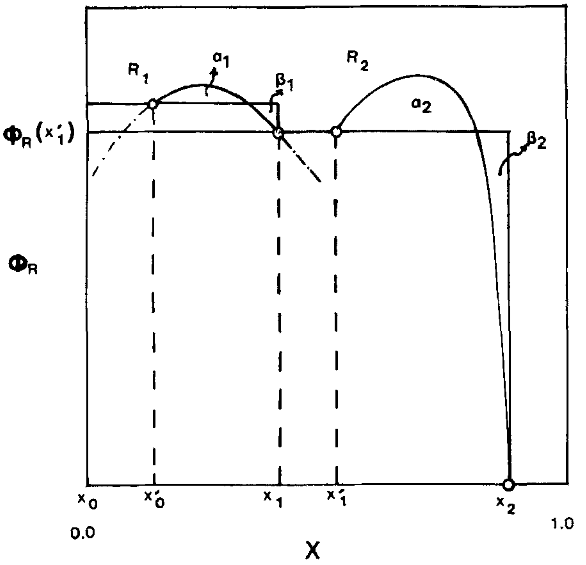


Figure 1. Graphical interpretation of necessary conditions for the optimality of a serial combination of two recycle reactors for a Type II complex reaction.

$$\frac{\partial p_1}{\partial x_1} = \phi_R \Big|_{x = x_1'} \tag{10}$$

From the definition of ϕ this can be written as

$$\phi_R \big|_{x = x_1} = \phi_R \big|_{x = x_1'} \tag{11}$$

From Eqs. 6 and 9 the following necessary condition is obtained for Type II reactions

$$\frac{\partial x_2}{\partial R_2}[(R_2 + 1)\phi_R(x_2) - R_2\phi_R(x_1')] - \phi_R(x_1')(x_2 - x_1)/(R_2 + 1) + \int_{x_1'}^{x_2} \phi_R dx = 0 \tag{12}$$

The sufficient conditions for maximum p_2 are

$$\frac{\partial^2 p_2}{\partial x_1^2} \leq 0 \tag{13}$$

$$\frac{\partial^2 p_2}{\partial R_2^2} \leq 0 \tag{14}$$

Equations 6 and 13 result in the following condition for Type II reactions

$$\frac{\partial^2 p_1}{\partial x_1^2} - \frac{\partial \phi_R}{\partial x_1'}\Big|_{x = x_1'} \frac{1}{(R_2 + 1)} \leq 0 \tag{15}$$

The necessary and sufficient conditions can be graphically shown in Figure 1 where the instantaneous yield ϕ has been plotted versus conversion, x. The necessary condition, Eq. 10, indicates that the value of the instantaneous yield at $x = x_1$ is equal to that at $x = x_1'$ as shown by the horizontal line in Figure 1. The last term in Eq. 12 is the area under the second curve from $x = x_1'$ to $x = x_2$. The second term is the area of the rectangle with sides $\phi_R(x_1')$ and $(x_2 - x_1)$, since from Eq. 7 $(x_2 - x_1)/(R_2 + 1)$ is equal to $(x_2 - x_1')$. The difference in the areas from Eq. 12 is given by:

(Area of region β_2 in Figure 1)

$$- \text{(Area of region } \alpha_2 \text{ in Figure 1)} = \frac{\partial x_2}{\partial R_2}[(R_2 + 1)\,\phi_R(x_2) - R_2\phi_R(x_1')] \tag{16}$$

In the case of nondegradable reaction products, the $\partial x_2/\partial R_2$ term in Eqs. 12 and 16 becomes insignificant, and according to Eq. 16 the area of region β_2 in Figure 1 becomes equal to the area of region α_2.

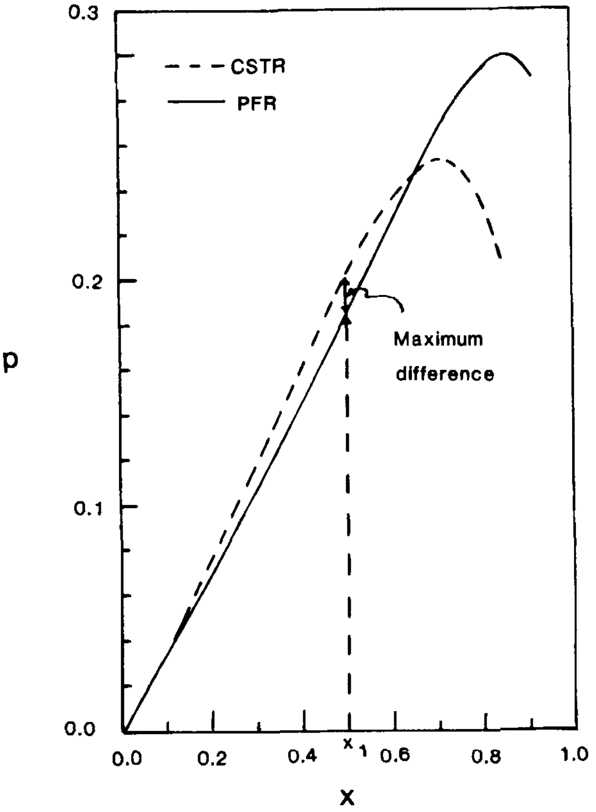


Figure 2. Graphical representation of maximum difference rule for the Van de Vusse reaction with a_1 = 2.0 and a_2 = 0.5.

It must be emphasized that Eqs. 11 and 12 or 16 are necessary conditions for local maximization of the product concentration, p_2. The absolute maximum may lie at the boundaries of the variables, such as at $R_2 = 0$ or $R_2 = \infty$. Hence, if a local maximum stationary point cannot be located to satisfy Eqs. 10 and 12, the limiting values at $R_2 = 0$ and $R_2 = \infty$ have to be compared to determine the optimal x_2.

Equations 6 through 16 can also be applied to the first recycle reactor and, with the necessary condition, for the maximization of product concentration, p_1. These conditions will similarly result in Eq. 16 where the difference in the area of regions β_1 and α_1,

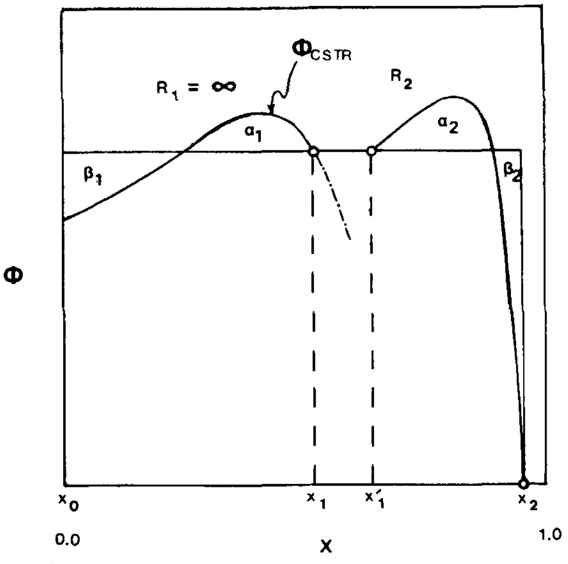


Figure 3. Graphical interpretation of necessary conditions for the optimality of a combination of CSTR followed by a recycle reactor for Type II complex reactions.

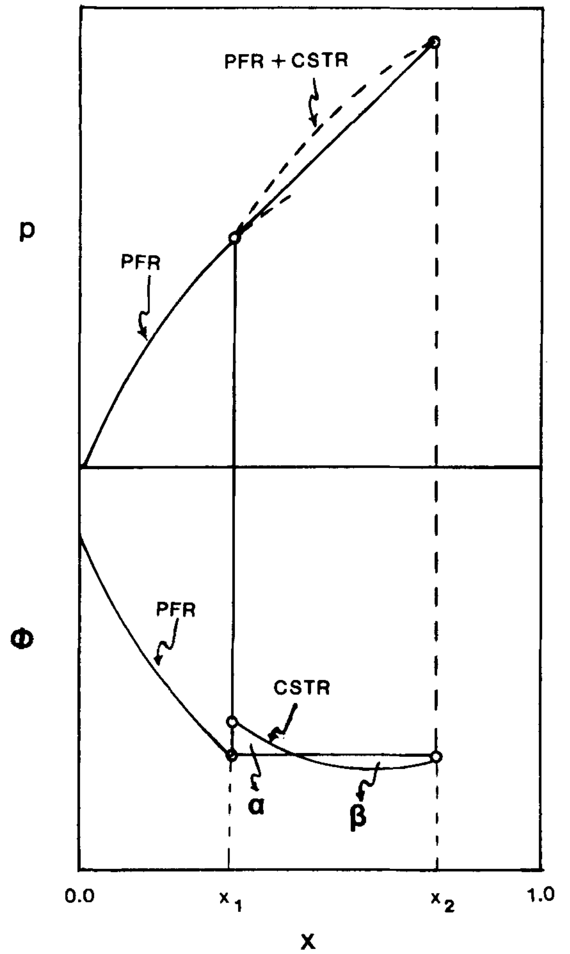


Figure 4. Graphical interpretation of necessary conditions for the optimality of a PFR followed by a CSTR for Type II complex reactions.

shown in Figure 1, is given by the right hand side of Eq. 16 written for the first reactor. This equation enables the point x_1 to be located in Figure 1.

When the first reactor is a CSTR (i.e., when $R_1 = \infty$), and the second reactor is a PFR (when $R_2 = 0$), Eqs. 8 and 10 reduce to Eq. 17 and Eqs. 12 and 14 reduce to Eq. 18:

$$\frac{\partial}{\partial x_1}(p_{1,\mathrm{CSTR}} - p_{1,\mathrm{PFR}}) = 0 \tag{17}$$

$$\frac{\partial^2}{\partial x_1^2}(p_{1,\mathrm{CSTR}} - p_{1,\mathrm{PFR}}) \leq 0 \tag{18}$$

If the product concentration, p, is plotted versus the conversion, x, for a single CSTR and a single PFR, as shown in Figure 2, the product concentration difference between the CSTR and PFR is maximum at the intermediate conversion, x, between the CSTR and PFR. This is the maximum difference rule that has been shown to give the optimum intermediate conversion (x_1) for a CSTR + PFR system for the Van de Vusse reaction scheme (Chitra and Govind, 1982).

On the instantaneous yield plot (ϕ vs. conversion, x) shown in Figure 3, note that the ϕ_{CSTR} curve must satisfy Eq. 12 or Eq. 16 for the CSTR to be the optimal first reactor. Dividing Eq. 16 by $(R_2 + 1)$ and taking the limit $R_2 \rightarrow \infty$ or $R_2/(R_2 + 1) \rightarrow 1$ and $x_1' \rightarrow x_2$, it is clear that the areas of regions α_1 and β_1 become equal. The regions α and β are indicated in Figure 3. The second reactor in Figure 3 is a recycle reactor, and the areas of regions α_2 and β_2 also satisfy Eq. 16. If the second reactor is a PFR, the points x_1 and x_1' coincide and the second ϕ vs. x curve is for a PFR.

When the first reactor is a PFR ($R_1 = 0$) and the second reactor is a CSTR ($R_2 = \infty$), Eqs. 6 and 13 reduce to the following:

$$\frac{p_2 - p_1}{x_2 - x_1} = \phi_{\mathrm{CSTR}}\Big|_{x = x_2} \tag{19}$$

and

$$\frac{\partial^2 p_2}{\partial x_1^2} \leq 0 \tag{20}$$

Equations 10 and 14 reduce to the following equations:

$$\frac{\partial p_{1,\mathrm{PFR}}}{\partial x_1} = \phi_{\mathrm{CSTR}}\Big|_{x = x_2} \tag{21}$$

and

$$\frac{\partial^2 p_1}{\partial x_1^2} \leq 0 \tag{22}$$

From Eqs. 19 and 21 we get

$$\frac{\partial p_{1,\mathrm{PFR}}}{\partial x_1} = \frac{p_2 - p_1}{x_2 - x_1} \tag{23}$$

Equations 21, 22, and 23 are shown graphically in Figure 4. The intermediate conversion x_1 between the PFR and CSTR is located at the point where the minimum occurs in the instantaneous yield function ϕ as given by Eq. 22. At this intermediate concentration, the slope of the product concentration curve, p vs. x, is shown in Figure 4 as a straight line tangent to the PFR curve. From Eqs. 20 and 22 the final exit conversion, x_2, must lie on this tangent and must also satisfy Eq. 21. The solution of Eqs. 21 and 23 determines the optimum final conversion, x_2.

The above rules can be extended to a serial combination of N reactors as shown in Figure 5. The complete procedure for obtaining the optimal reactor structure for Type II reactions is as follows (Figure 6).

1. Select a value for the recycle ratio for the first reactor, R_1. Plot the instantaneous yield, ϕ_R, versus the conversion x for the initial boundary conditions $x_0 = 0$, $p_0 = 0$.
2. Select a point, a, on the curve (Figure 6) such that the difference in the areas for regions α_1 and β_1 satisfies Eq. 16.
3. Guess a value for recycle ratio R_2 for the second reactor and plot the ϕ_R curve.
4. The intersection point, b, is located by drawing a horizontal line from point a, as shown in Figure 6.
5. Calculate point c from the material balance equation

$$x_2 = \frac{x_1'(R_2 + 1)}{R_2} - \frac{x_1}{R_2}$$

6. Check whether Eq. 16 is satisfied for regions α_2 and β_2.
7. If Eq. 16 is not satisfied, select another value for R_2 and repeat steps 3 through 6.
8. Repeat steps 3 through 7 until the final conversion x_n is obtained.

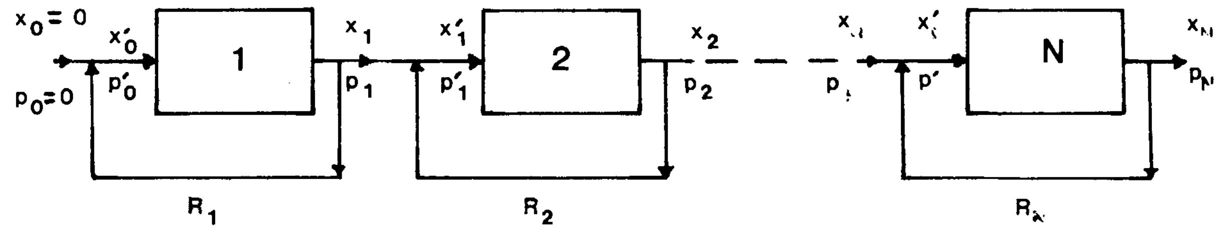


Figure 5. Generalized serial isothermal reactor configuration consisting of recycle reactors.

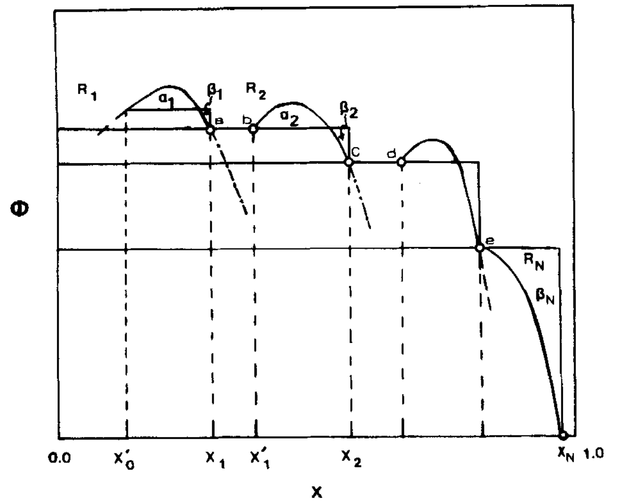


Figure 6. Graphical procedure for obtaining optimum serial isothermal reactor configuration for Type II complex reactions.

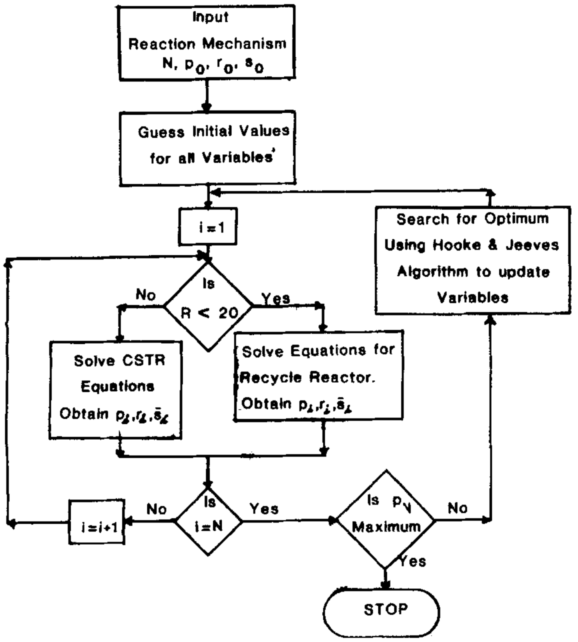


Figure 7. Overall procedure to obtain the optimum serial isothermal reactor configuration for Type III complex reactions.

9. Repeat steps 1 through 8 for different values of R_1 and select the value for R_1 which will maximize the final product concentration, p_N.

Figure 5 is not completely general. For example, a recycle stream from a downstream reactor to an upstream reactor cannot be represented by the serial configuration. Nonserial structures for two reactors are qualitatively discussed in a later section.

For Type III complex reactions, it is not possible to derive analytical results which would apply in general. However, for a serial reactor structure as shown in Figure 5, the final product concentration, p_N, can be written as

$$p_N = fn[p_0,r_0,\bar{s}_0,(p_i,r_i,\bar{s}_i,R_i,i=1\ldots N)] \tag{24}$$

The final concentration, p_N, can be maximized by using the Hooke and Jeeves (1961) optimization algorithm. The overall procedure is shown in Figure 7. A computer program has been developed based on this procedure to derive the optimum serial reactor configuration for any Type III complex reaction.

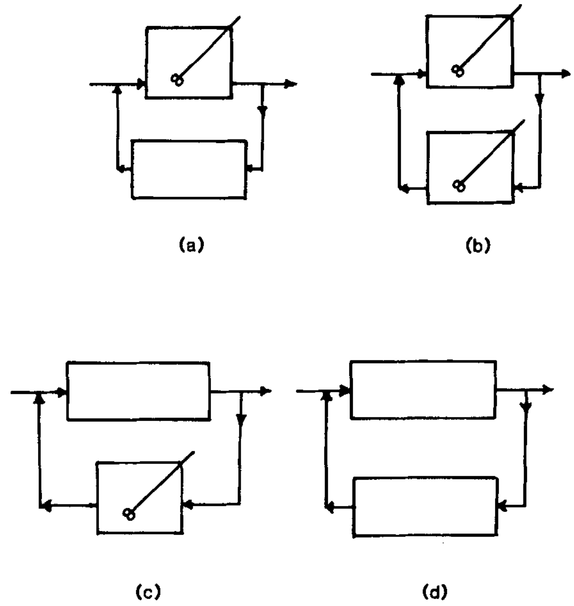


Figure 8. Some non-serial reactor configurations with recycle for isothermal operation.

NON-SERIAL CONFIGURATIONS

For isothermal reactors, parallel combinations will not improve the product yield but simply increase the productivity relative to a single reactor. Reactor combinations with recycle can improve the product yield and some are shown in Figure 8. Structures (*a*) and (*b*) will not improve the yield since the CSTR in itself is a completely mixed reactor. Structure (*c*) is not optimum for degradation reaction since mixing at the end of the reacting unit (PFR followed by CSTR) will not improve the product yield. The structure may be optimum for reactions where mixing at the end of the reacting unit will improve the yield. Structure (*d*) may be optimum for reactions where the mixing of the reaction mixture is desired without incurring product degradation.

SYNTHESIS PROCEDURE FOR SERIAL ISOTHERMAL REACTOR STRUCTURES

The synthesis procedure begins by the user specifying the reaction mechanism, the kinetic constants, and the feed composition. Once the product and the limiting reactants are specified, the synthesis program classifies the input reaction into one of three types as described earlier in this paper. If the reaction is a Type I (simple) reaction the existing rules proposed by Levenspiel (1962), Denbigh (1961), and others can be used to specify the reactor type and structure. If the reaction is a Type II complex reaction, the graphical procedure presented in this paper can be used to find the optimal reactor structure. However, for a Type III complex reaction, the computer program using the optimization procedure can be used to find the optimal structure. Once the reactor type and structure have been specified, the program calculates the required reactor volumes.

Example 1.

Consider the following Van de Vusse (1964) reaction

$$A \xrightarrow{k_1} B \xrightarrow{k_2} C$$

$$A + A \xrightarrow{k_3} D$$

The reactant is A and product is B. The reaction rate constants are:

$$k_1 = 2\ \text{h}^{-1}$$

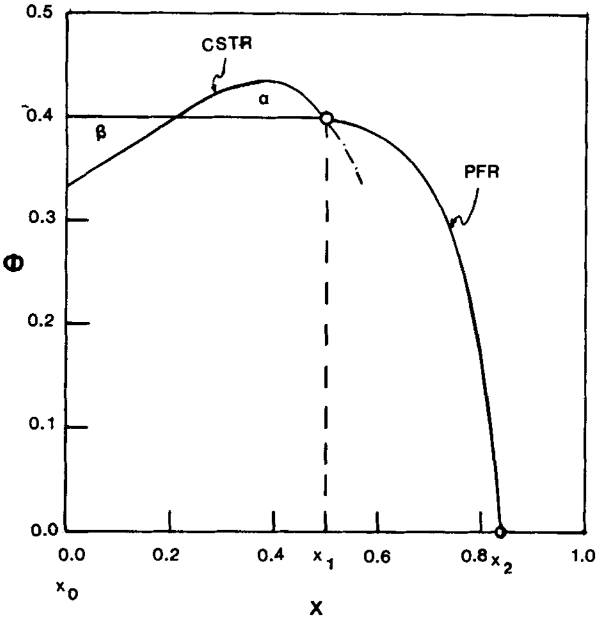


Figure 9. Graphical procedure to obtain the optimum reactor combination for the Van de Vusse reaction with a_1 = 2.0 and a_2 = 0.5.

$$k_2 = 1\ \text{h}^{-1}$$

$$k_3 = 1\ \text{L} \cdot \text{gmol}^{-1} \cdot \text{h}^{-1}$$

with a feed (pure A) flow rate of 100 L/h and initial concentration of reactant A equal to 4.0 gmol/L. The instantaneous yield function for this reaction and for a single recycle reactor can be written as

$$\phi_R = -\frac{dp}{dr} = \frac{r - a_2 p}{r + a_1 r^2} \tag{25}$$

where
$r = A/A_0$ dimensionless concentration of reactant A
$p = B/A_0$ dimensionless concentration of product B
$a_1 = k_3 A_0/k_1$ dimensionless kinetic parameter
$a_2 = k_2/k_1$ dimensionless kinetic parameter

with the following reactor inlet conditions

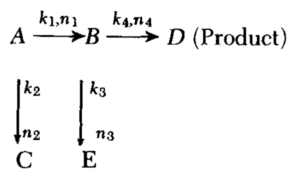
$$r_0' = \frac{r_0 + R_1 r_1}{R_1 + 1} \tag{26}$$

$$p_0' = \frac{p_0 + R_1 p_1}{R_1 + 1} \tag{27}$$

Since the rate of product formation is a function of reactant and product concentrations only, this reaction is a Type II complex reaction. Based on the procedure derived for synthesizing the serial reactor configurations, a CSTR + PFR serial combination is found to be optimal. The graphical procedure for the optimal reactor system is shown in Figure 9. Note that the areas of regions α and β are equal for the first reactor (CSTR), which satisfies Eq. 16. For the second reactor, Eq. 16 cannot be satisfied and the boundary value of $R_2 = 0$ (PFR) is found to be optimum. This result can also be obtained by the maximum difference rule (Chitra and Govind, 1982), given by Eqs. 17 and 18. Note that the intermediate reactant conversion between the CSTR and PFR is the point at which the maximum difference occurs in the product concentrations between a single CSTR and a single PFR as shown in Figure 2. From the instantaneous yield plot, shown in Figure 9, at the maximum difference value of conversion the area rule from Eq. 16 is also satisfied.

Example 2.

Consider the following Denbigh reaction



$$\begin{tikzcd} A \arrow[r, "k_1{,}n_1"] \arrow[d, "k_2" ', "n_2"] & B \arrow[r, "k_4{,}n_4"] \arrow[d, "k_3", "n_3" '] & D\ \text{(Product)} \\ C & E & \end{tikzcd}$$

The desired product is D. The reaction rate constants and reaction orders are

$k_1 = 1\ \text{L} \cdot \text{gmol}^{-1} \cdot \text{s}^{-1}$ $n_1 = 2$
$k_2 = 0.6\ \text{s}^{-1}$ $n_2 = 1,$
$k_3 = 0.1\ \text{L} \cdot \text{gmol}^{-1} \cdot \text{s}^{-1}$ $n_3 = 2$
$k_4 = 0.6\ \text{s}^{-1}$ $n_4 = 1.$

The initial feed (pure A) concentration is 6 gmol/L. The instantaneous yield function for this reaction mechanism and for a single recycle reactor can be written as

$$\phi_R = -\frac{dp}{dr} = \frac{b_4 s}{r^2 + b_2 r} \tag{28}$$

$$-\frac{ds}{dr} = \frac{r^2 - b_4 s - b_3 s^2}{r^2 + b_2 r} \tag{29}$$

where

$r = A/A_0$ dimensionless concentration of reactant A
$p = D/A_0$ dimensionless concentration of product D
$s = B/A_0$ dimensionless concentration of intermediate species B
$b_2 = k_2/(k_1 A_0)$
$b_3 = k_3/k_1$ dimensionless
$b_4 = k_4/(k_1 A_0)$ kinetic parameters

with the following reactor inlet condition

$$r_0' = \frac{r_0 + R_1 r_1}{R_1 + 1} \tag{30}$$

$$s_0' = \frac{s_0 + R_1 s_1}{R_1 + 1} \tag{31}$$

$$p_0' = \frac{p_0 + R_1 p_1}{R_1 + 1} \tag{32}$$

Since the above equation clearly indicates that the rate of product formation is a function of reactant concentration and the intermediate species B concentration, this reaction is a Type III complex reaction. Since in this reaction mechanism, degradation of the desired product D does not occur, maximum reactant conversion was specified as 95%. Using the computer program developed based on the overall procedure shown in Figure 7, a serial combination of a PFR + CSTR is found to be the optimum configuration. The final product concentration is 2.92 gmol/L for a feed flow rate of 10 L/s. The volume of the plug flow reactor is 42.3 L and that of the CSTR is 73.3 L.

NOTATION

a_1, a_2	= dimensionless kinetic parameters, Example 1
b_1, b_2, b_3, b_4	= dimensionless kinetic parameters, Example 2
f_1, f_2, f_3, f_4, f_5	= reaction rate functions, Eqs. 1, 2, 3, 4, and 5.
k_1, k_2, k_3	= reaction rate constants for Van de Vusse reaction, Example 1
$k_1', k_2', k_3' k_4'$	= reaction rate constants for Denbigh reaction, Example 2
n	= number of other species, Eqs. 1 and 2
N	= number of reactors in the structure
p	= dimensionless product concentration, Eqs. 1 and 2
$p_0, p_1 \ldots p_N$	= dimensionless product concentrations at

different locations of the reactor train, Figure 5

P = product concentration

r = dimensionless limiting reactant concentration, see Eqs. 1 and 2

$r_0, r_1 \ldots r_N$ = dimensionless limiting reactant concentration at different locations of the reactor train, Figure 5

R = limiting reactant concentration

$R_1, R_2 \ldots R_N$ = recycle ratio; ratio of volumetric recycle stream flow rate to the volumetric feed rate for different reactors in the reactor train

R_0 = initial limiting reactant concentration

$\bar{s} = (s_1, s_2 \ldots s_i \ldots s_n)$ = vector of dimensionless concentration of other species participating in the reaction

$s_i = \frac{S_i}{R_0}$ = dimensionless concentration of other (intermediate, by-product, nonlimiting reactant) species S_i

S_i = concentration of other species

$\bar{s}_0$ = vector of initial dimensionless concentration of other species

x = $1 - r$, conversion of the limiting reactant

$x_0, x_1 \ldots x_N$ = conversion of the limiting reactant at different locations of the reactor train, Figure 5

Greek Letters

α = area under the instantaneous yield curve but above the rectangle, Figure 1

$\alpha_1, \alpha_2 \ldots \alpha_N$ = α values for different reactors in the reactor train

β = area under the rectangle but above the instantaneous yield curve, Figure 1

$\beta_1, \beta_2 \ldots \beta_N$ = β values for different reactors in the reactor train

ϕ = instantaneous yield of the product

ϕ_R = instantaneous yield of the product for the recycle reactor

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Manuscript received Feb. 16, 1983; revision received Oct. 11, 1983, and accepted Oct. 20.