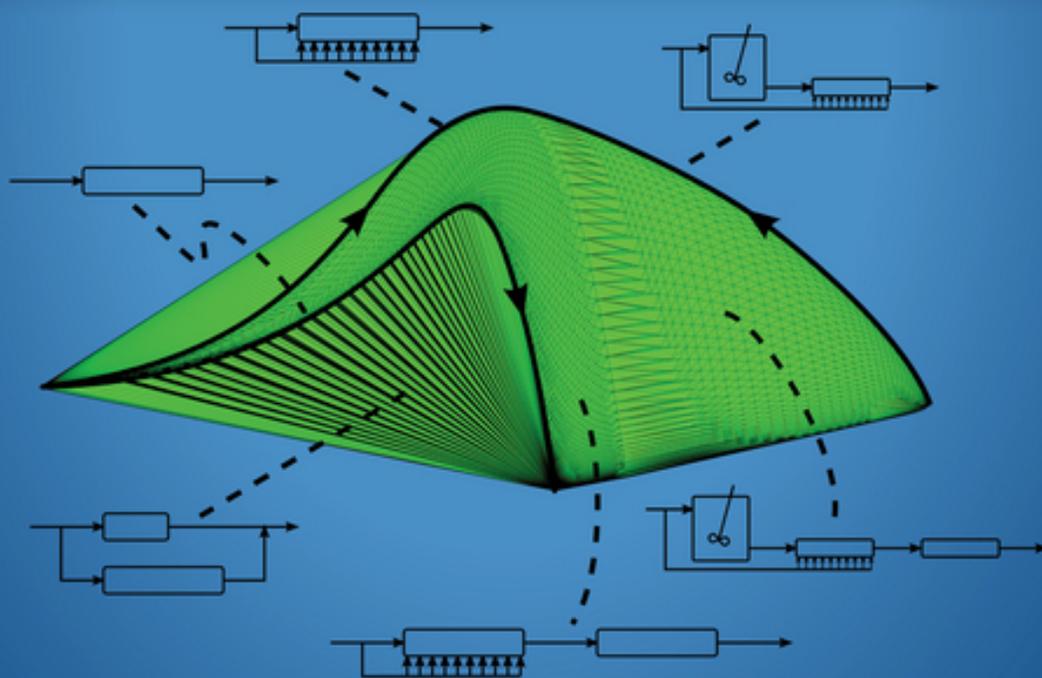


ATTAINABLE REGION THEORY

AN INTRODUCTION TO CHOOSING AN OPTIMAL REACTOR



DAVID MING | DAVID GLASSER | DIANE HILDEBRANDT
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An Introduction to Choosing an Optimal Reactor

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PREFACE

The problem that this book sets out to solve can be formulated very simply. Given a set of chemical reactions with known kinetics, what is the best reactor that can be used to carry out the set of reactions. As easy as the question is to ask, the answer is not obvious, as on the face of it there are an infinite number of possibilities. What this preface will do is to outline the 50-year journey to solve this problem. This history also embodies a cautionary tale for those doing (and wanting to do) research.

I did my PhD looking at a reaction and its kinetics. As a result of this, I became interested in chemical reactor theory. I remember, in particular, the excellent books of Denbigh and Levenspiel. Both of them talked about the aforementioned problem and documented some heuristics to help industrialists to come up with some solutions. However, it was recognized that these heuristics were sometimes contradictory.

At that stage, I became interested in optimization in general. Also, on my first sabbatical leave, I was able to work with some of the greats of chemical engineering (Stanley Katz, Reuel Shinnar, and Fritz Horn) and became involved solving some more limited optimization problems, such as using contact times for catalytic reactions, and minimizing holding times for a series of continuous-flow stirred tank reactors (CSTRs). Also it was at this time that I became interested in using Pontryagin's maximum principle (developed for space exploration problems) on chemical reactor problems. This was the stage that I, at first, thought we could solve the main problem by extending residence time distribution theory to nonlinear kinetics. There was the well-known result for segregated systems that one could work out conversions for all kinetics using the residence time distribution. I thought we should be able to extend this theory for all possible reactors and then use the maximum principle to solve the general problem.

On the next two sabbaticals, I worked with Roy Jackson and Cam Crowe to take this idea further, and in the end, much

to our joy, Roy Jackson and I solved the problem (after 15 years!). The joy soon evaporated as we realized that even though we had a complete description for all possible reactors with nonlinear kinetics, in order to solve the main problem we had to find a function that could have a non-countable infinity of changes with possibly some of the values going to infinity. This was clearly an impossible problem to solve.

This meant going back to the drawing board! I started to fiddle with some simple problems from the literature by drawing simple two-dimensional graphs. What I soon realized is that if there were concavities on the graphs, they could be filled in with straight lines to make what are called convex hulls. What was really exciting was that these lines were nothing more than mixing between two points on the graph. Out of this came the idea that a reactor was a system that was made up of two processes: reaction and mixing. Each of these processes can be represented on a graph as vectors. Suddenly, the problem I was looking at changed to a geometric one, as I was now looking at making the region in the space as large as possible using the two vector processes. When visiting Martin Feinberg at Rochester, he drew my attention to a paper by Fritz Horn that talked about what he termed attainable regions (ARs). That is, in this context, the largest region in some component space that one could obtain using any processes. He showed that if one had this AR, the optimization problem was relatively simple. Without at first realizing what I was doing, I had found a geometric way of finding the AR for reacting systems. At this stage, I was only doing it graphically on problems that could be represented in two dimensions.

At this stage, it became clear why one could not solve this problem using standard optimization methods. This was because mixing is not a differential process in the ordinary sense. That is, one could not use methods that only looked in the immediate neighborhood as one could mix from any point that was itself attainable, and this could be far

from the neighborhood of the point we were examining. An interesting point is that the AR method can solve these non-continuous problems, and so is essentially a new method of optimization, but to the best of my knowledge this idea has not been taken up in other fields of study.

When I arrived back from sabbatical, I looked for a student to work on the problem to extend the results. At this stage, Diane Hildebrandt, who I had taught as an undergraduate, and had done her MSc with me, was looking for a PhD project. I warned her that the project was a little open-ended. However, she was sufficiently interested in the topic (was this brave or foolhardy?) to want to work on it. In the end, out of her work, came the foundations of our work on AR theory. At last, we had a method that could solve the problem I had started out to look at more than 20 years earlier.

What is really interesting (and important) is that we were able to solve a problem that was generally regarded as impossible by thinking about it in a very different way. Instead of trying to answer the problem of finding the optimal reactor in one step, we had first asked what processes are occurring in a chemical reactor, how can we represent them, and how can we then use these processes to ask what are all possible outcomes. Only at this stage do we look at how to perform the optimization. By breaking the problem up into smaller stages, in the end, we were able to solve it.

Publishing the first AR paper turned out to be a bit of a nightmare! As reviewers knew solving this problem was not possible, faith even in scientists was more important than reason. The paper kept being rejected with reviewers telling us there must be something wrong with what we were doing, even though they could not say what it was. In the end, we had to say to the editor if the reviewers could not find the mistake, he should publish it and wait for someone to come up and show us the fallacy. Luckily, in the end he agreed, and we are still waiting for someone to show us why what we did was wrong!

At this stage, my son Benjamin, also a chemical engineer, worked on AR theory for his MSc, for the first industrially interesting problem. Diane then went on sabbatical where she worked with Martin Feinberg to formalize some of the ideas she had started on her PhD. Out of this arose the idea that there are only three types of reactor needed to find the AR in any dimensions, namely plug flow reactors (PFRs), continuous-flow stirred tank reactors (CSTRs), and differential sidestream reactors (DSRs). Suddenly, from there being an infinite number of possible reactors needed in an optimal system, there are only three types. The significance of this is that while initially we needed to find the AR before doing the optimization, it is now, in principle, possible to generate an algorithm to solve large problems without necessarily finding the full AR, which may potentially be a very large problem in higher dimensions.

The method became a topic of research for us, and some others, and we published many papers in the area. Also the

method became accepted and now appears as standard in many textbooks on chemical reactor theory and design. In particular, we began to look at ways of calculating ARs in higher dimensions in order to be able to have a successful algorithm that could be used fairly generally. David Ming has worked on this and also extended the ideas from steady-flow reactors to batch reactors. Matt Metzger who spent time with us from the United States found the topic interesting and helped to gather all the information in a logical and consistent way and helped to see that it could be presented in a readable way.

Thus, even though the story at this stage is complete, I believe there is a lesson we can learn from this. I call this the four P's of research. These are as follows:

1. Passion
2. Patience
3. Perseverance
4. Persuasion

I believe the history of this book typifies all of these aspects.

PASSION

If I (and others) had not believed that this project was worthy of spending time on it, it would not have been done.

PATIENCE

I worked patiently on it for 15 years to get an answer that was of no real use.

PERSEVERANCE

Perseverance is the hard work you do after you get tired of doing the hard work you already did. —Newt Gingrich. I believe this speaks for itself in the context of this work.

PERSUASION

It is not sufficient to do good work on its own; one needs to persuade the rest of the world. We really struggled with this! Thomas Grey said it all.

*Full many a gem of purest ray serene,
the dark unfathomed caves of ocean bear,
full many a flower is born to blush unseen
to waste its sweetness on the desert air*
David Glasser

ACKNOWLEDGMENTS

The dilemma that we face with attainable region (AR) theory is that some aspects of the field are very easy to grasp, whereas other parts are very difficult. Over the years, we have done our best to accommodate for these different aspects, yet there has not been a single, consistent, treatment of AR theory that could be used to address multiple audiences and interest groups. It is our hope that this textbook will not only provide this consistency, but it will also be used by those who wish to understand the field of AR theory for the first time.

The opportunity to write a textbook comes with a mix of excitement and trepidation. On one hand, it is exciting to be given an opportunity to share our ideas—many of which have been developed over *many* years by *many* people—and to articulate them in our own way. On the other hand, it is daunting to think of how best to do this, so that we live up to the expectations of all those who have contributed to these ideas, and also to those prospective readers who are coming to the field for the first time. Indeed, attempting to write a coherent set of work, suitable for many points of view, has been a challenging task, and this textbook would not exist without the help and support from our family, friends, and colleagues who have helped at various stages along the way. In particular, we are very grateful to Cody Bishop and Kwame Owusu-Ansah for helping to proofread (and correct) early versions of the work. We would also like to thank Michelle Low, not only for supporting us throughout

the course of this work but also for contributing her time and creativity to take many of the excellent photographs used in this book. Your diligence and continuous enthusiasm throughout this time has been invaluable to us. We are also grateful to Professor Arun Moharir for helping to supply and organize many of the examples and ideas for the companion website. We thank Linda Anthony and Eric Lam, for allowing much of the early stages of this work to be developed at Rutgers University.

Last, we thank our family and friends who have supported and encouraged us throughout this journey. Thank you for understanding and also for sometimes *not* understanding, but still supporting us! In the book, we try to emphasize a common theme of *you can't fix what you don't know*, as a way to explain the importance of understanding performance bounds. In many ways, this theme is mirrored by your support. You have helped us to understand our own limits, and your encouragement has made this task much more achievable. Thank you.

DAVID MING
DAVID GLASSER
DIANE HILDEBRANDT
BENJAMIN GLASSER
MATTHEW METZGER

PRIOR KNOWLEDGE

This book assumes a basic understanding of chemical reaction stoichiometry and material balances, equivalent to that administered in an undergraduate course in chemical engineering or chemistry. Knowledge of introductory chemical reactor theory is also beneficial, although it is not a requirement.

We shall be interested in a broad, macroscopic, view of reactors—one where elementary mass balances are applied over entire process units. That is, we are not interested in *modeling* the system in detail, and attainable region (AR) theory does not demand the use of microscopic transport equations for instance.

AR theory has a *geometric* flavor. Hence, readers who possess a basic understanding of calculus (derivatives and integration) and linear algebra (vectors and matrices) may find the introductory concepts of AR theory easier to follow. Familiarity with plotting graphs in two and three dimensions on a computer is also helpful. Nevertheless, we shall always strive to provide the necessary theory and tools, when appropriate. This fulfills the dual purpose of introducing the reader to the particular terminology and nomenclature adopted in this book.

HOW THIS BOOK IS STRUCTURED

The material covered in this book is organized into two sections. It may be helpful to refer to Figure P.1 for an overview of the organization of chapters. Section I (Chapters 1–5) focuses on the basics of attainable region (AR) theory. Importantly, this section introduces a different way of viewing chemical reactors and reactor networks. The examples discussed in Section I are of a simpler nature, with an emphasis on describing all problems in two dimensions only. Section I is best read in a sequential fashion.

Section II (Chapters 6–10) focuses on Extended topics in AR theory, and also discusses specific fields of research that may be of interest to select readers. These chapters are more mathematical in nature, and may appeal to readers with a stronger background in computer programming and mathematical modeling. Whereas Section I is intended for senior undergraduates, Section II is more suited for graduate

students and working professionals. We hope that these chapters will be used to gain a deeper appreciation for what is an exciting field of study in chemical reactor network synthesis. Although there is more flexibility in how this section can be tackled, we suggest reading Chapters 6 and 7 together.

Chapter 1: In this chapter, we introduce the idea of the reactor network synthesis problem and performance targeting. Ultimately, we attempt to articulate two important messages: *how do you know you have achieved the best? You cannot fix what you don't know.*

Chapter 2: In this chapter, we introduce the idea of concentration and mixing from a geometric perspective. We look at how data may be interpreted in the phase plane (concentration space) and the link between mixing, convex hulls, and attainability. Readers already familiar with vectors, and the

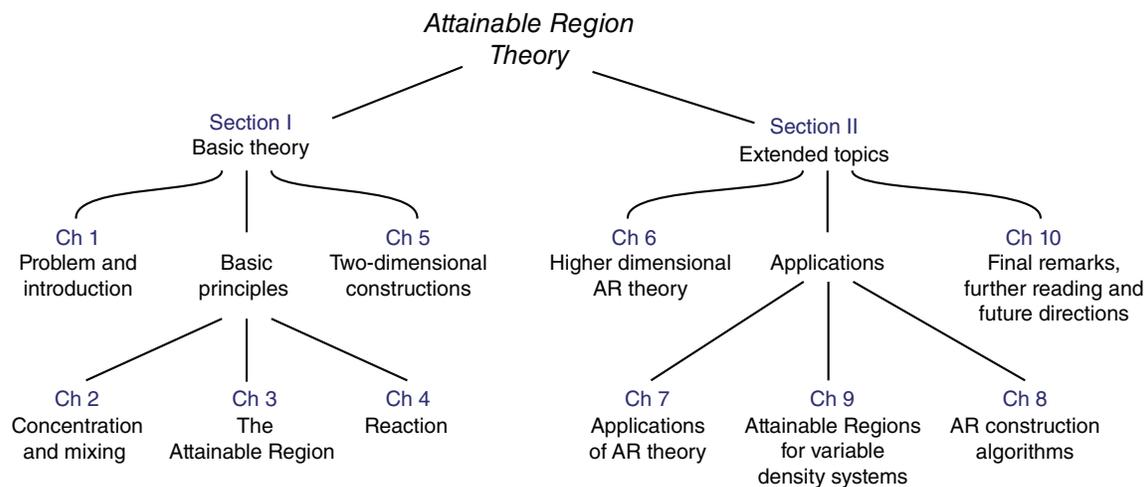


Figure P.1 Organization of chapters.

rudiments of ordinary differential equations, may choose to skip Chapter 2 and move on to Chapter 3, where the AR is described.

Chapter 3: In this chapter, we apply the concepts developed in Chapter 2 to a simple thought experiment and show how system performance may be improved with these ideas. This leads us to introduce, for the first time, the idea of the AR for a reactive system. Basic properties of the AR are also discussed.

Chapter 4: In this chapter, we discuss how reaction may also be viewed from a geometric perspective. We introduce the three fundamental reactor types used in AR theory, and we also discuss additional properties of the AR related to reaction.

Chapter 5: In this chapter, we look to apply our knowledge of concentration, reaction, and mixing to simple systems. We generate the AR for a number of two-dimensional systems and show how the AR approach can lead to insights into the design of chemical reactor networks.

Chapter 6: In this chapter, we describe the nature of the AR boundary in higher dimensional spaces, and we explain a number of important results used in AR theory. We also look at ways of calculating conditions to reside on the AR boundary in any dimension.

Chapter 7: In this chapter, we discuss a number of worked examples for more complex systems, including higher dimensional systems and constructions where there is an equipment constraint. We also discuss how AR theory can be applied to batch reactors.

Chapter 8: In this chapter, we describe a number of AR construction algorithms that may be implemented on a computer.

Chapter 9: In this chapter, we look at how to extend concepts of the AR to variable density systems, such as non-ideal systems in the gas phase.

Chapter 10: We conclude by providing summaries and key take-away points from each chapter. This chapter also provides a small set of literature for further reading as well as potential research interests.

ICONS

Throughout the book, look out for icons to signal an interesting or important aspect of the text. These could be worked examples, important ideas, or simply extra information.



CONCEPT: An idea that may be useful in later discussions.



EXAMPLE: A question, or set of questions, that are intended to test your understanding of an idea, or to extend on a specific topic.



ILLUSTRATION: A worked example or discussion demonstrating an application of AR theory or an extension of an idea.



IMPORTANT: An important concept that should be kept in mind.



OBSERVATION: An idea or a result that may not be obvious that we wish to highlight.



RECALL: A concept or relation developed previously that we wish to highlight again.



SIDE NOTE: Additional information or an interesting fact that is related to the present discussion.

SOFTWARE AND COMPANION WEBSITE

Due to the geometric nature of attainable region (AR) theory, as well as the complexity of the systems considered, we often need software tools to help us interpret and visualize our problems. Rather than attempting to populate a conventional CD-ROM with software and additional examples that cannot be adapted over time, we have decided to release this material on a companion website (<http://attainableregions.com>), which has been developed for this book. We hope that this approach will allow us to cater to the changing needs of the reader and AR community as a whole, where these software resources can be tailored accordingly over time.

Figures and demonstrations supplied in the book have been produced primarily in MATLAB. However, the use

of MATLAB is not a requirement for understanding the examples in this book or using the companion software on the website. Instead, Python (specifically the Anaconda distribution) has been chosen for the website examples and demonstrations. Python may be used without any restriction, even for commercial use. It is freely available on the Internet, and, importantly it is also well supported within the scientific and engineering community. Information on how to install the Anaconda distribution and how to access the website examples can be found both in Appendix C, and on the companion website.

NOMENCLATURE

Commonly used symbols and notation adopted in this book are listed in the following table. Symbols that are only used once for a particular description and that are defined at the point of use may not be listed. Also note that some symbols may be used in different chapters for different situations, and thus more than one definition may apply.

VECTOR, MATRICES, AND SETS

A	Stoichiometric coefficient matrix for a system of reactions	
C	Molar concentration vector	$\mathbf{C} = [c_A, c_B, \dots, c_n]^T$
C⁰	DSR sidestream mixing concentration	
C₀	A point residing on a hyperplane H(n , b)	$\mathbf{b} = \mathbf{n}^T \mathbf{C}_0$
C*	Concentration vector from a mixture	$\mathbf{C}^* = \lambda \mathbf{C}_1 + (1 - \lambda) \mathbf{C}_2$
C[#]	Equilibrium concentration achieved in a reactor	
E	DSR controllability matrix	$\mathbf{E} = [\mathbf{K}, \mathbf{N}]^T$
F	Set of feed concentrations	$\mathbf{F} = \{\mathbf{C}_{f1}, \mathbf{C}_{f2}, \dots, \mathbf{C}_{fN}\}$
G	Mass flow rate vector for variable density systems	$\mathbf{G} = [g_1, g_2, \dots, g_n]^T$
I	The identity matrix	
J(C)	Jacobian matrix of r(C) . Sometimes also given as dr(C)	

L	Set of points belonging to a lineation	
K	Controllability matrix for a critical DSR excluding null space	
N	Null space of the stoichiometric coefficient matrix A	$\mathbf{N} = \text{null}(\mathbf{A}^T)$
n	Hyperplane normal vector	
n_i	Molar flow rate vector	$\mathbf{n}_i = [c_{Ai}, c_{Bi}, \dots, c_{ni}]^T$
z	Mass fraction vector	$\mathbf{z} = [z_A, z_B, \dots, z_n]^T$
r(C)	Rate vector of a function of C	$\mathbf{r}(\mathbf{C}) = [r_A(\mathbf{C}), r_B(\mathbf{C}), \dots, r_n(\mathbf{C})]^T$
S	Set of points representing the stoichiometric subspace	
SX	The complement region of X to S .	
t(C)	CSTR tangent vector	$\mathbf{t}(\mathbf{C}) = [\mathbf{I} - \tau \mathbf{J}(\mathbf{C})]^{-1} \mathbf{r}(\mathbf{C})$
v(C)	Mixing vector	$\mathbf{v}(\mathbf{C}) = \mathbf{C}_2 - \mathbf{C}_1$
W	Molecular weight matrix	
X	Set of points representing a current set of achievable states	
z^[k]	The <i>k</i> th iterated Lie bracket	
ε	Extent of reaction vector	$\boldsymbol{\varepsilon} = [\varepsilon_1, \varepsilon_2, \dots, \varepsilon_n]^T$
0	The zero vector	$\mathbf{0} = [0, 0, \dots, 0]^T$

LETTERS

d	Number of independent reactions	
c_i	Molar concentration of component i	
$F(t)$	Fed-batch feeding rate as a function of reaction time	
G	Total mass flow rate through a reactor or reactor network	$G = \sum g_i$
g_i	Mass flow rate of stream i in a network	
$H(\mathbf{n}, b)$	Hyperplane defined by normal vector \mathbf{n} and scalar b	
k_i	Rate constant i in a rate expression	
M	Total mass	$M = \sum m_i$
m_i	Mass of component i	
n	Number of components participating in a reaction	
P	Reactor pressure	
P_k	Polytope representing the AR at iteration k	
Q	Volumetric flow rate through a reactor or reactor network	
$r_i(\mathbf{C})$	Rate expression for component i as a function of \mathbf{C}	
T	Reactor temperature	
t	Batch reaction time	
V	Reactor volume or total volume of a reactor network	
$V(t)$	Fed-batch volume as a function of reaction time	
W_i	Molecular weight of component i	
W_{avg}	Average molecular weight of a mixture	
Y_i	Yield of component i	
Z	The compressibility constant for real gases	$P\hat{V} = ZRT$
z_i	Mass fraction of component i	
\mathbb{R}	The set of all real numbers	
\mathbb{R}^n	The set of all vectors (n -tuples) with n real numbers (n -dimensional space)	

GREEK SYMBOLS

α	DSR sidestream mixing parameter for constant density systems
β	DSR sidestream mixing parameter for variable density systems

ε_i	Extent of reaction for reaction i in a system of reactions.	
θ	Angle between two vectors	$\mathbf{x}^T \mathbf{y} = \ \mathbf{x}\ \ \mathbf{y}\ \cos(\theta)$
$\Lambda(\mathbf{C})$	Critical CSTR determinant surface	
λ	Mixing fraction	
ν_i	Stoichiometric coefficient of a component i in a reaction	
ρ_{avg}	Average density of a mixture (by mass)	
σ	Reactor residence time in mass fraction space	$\sigma = V/G$
Σ_{ij}	Instantaneous selectivity of component i to j	
τ	Reactor residence time (constant density)	$\tau = V/Q$
Y_i	Instantaneous yield of component i	
$\varphi(\mathbf{C})$	vDelR critical DSR condition for a system in \mathbb{R}^3 ($d = 3$)	$\varphi(\mathbf{C}) = [\mathbf{J}(\mathbf{C})\mathbf{v}(\mathbf{C})]^T [\mathbf{v}(\mathbf{C}) \times \mathbf{r}(\mathbf{C})]$

MATHEMATICAL OPERATIONS AND ABBREVIATIONS

∇	Del operator (nabla)	$\nabla = \left[\frac{\partial}{\partial c_A}, \frac{\partial}{\partial c_B}, \dots, \frac{\partial}{\partial c_n} \right]^T$
$\ \mathbf{x}\ $	The norm of a vector \mathbf{x}	$\ \mathbf{x}\ = \sqrt{x_1^2 + x_2^2 + \dots + x_n^2}$
$L_f h(\mathbf{C})$	Lie derivative of a scalar function $h(\mathbf{C})$ with respect to a vector function $\mathbf{f}(\mathbf{C})$.	
$[\mathbf{f}, \mathbf{g}](\mathbf{C})$	Lie bracket of vector functions $\mathbf{f}(\mathbf{C})$ and $\mathbf{g}(\mathbf{C})$	$[\mathbf{f}, \mathbf{g}](\mathbf{C}) = d\mathbf{g}(\mathbf{C})\mathbf{f}(\mathbf{C}) - d\mathbf{f}(\mathbf{C})\mathbf{g}(\mathbf{C})$
$\text{Det}(\mathbf{A})$	Determinant of a matrix \mathbf{A}	
$\text{conv}(\mathbf{X})$	Convex hull of a set of points \mathbf{X}	
$\text{cl}(\mathbf{X})$	The closure of a set of points \mathbf{X}	
$\text{rank}(\mathbf{A})$	Rank of matrix \mathbf{A}	
$\text{null}(\mathbf{A})$	Null space of matrix \mathbf{A}	

SUBSCRIPTS

f	Refers to feed streams	$\mathbf{C}_f = [c_{Af}, c_{Bf}, \dots, c_{nf}]^T$
i, j, k, n	General reference to elements in a list, or index counter	

SUPERSCRIPTS

T	Matrix transpose	
#	Relating to an equilibrium state	$C^\#$ is an equilibrium concentration
*	Relating to a mixture	C^* is a concentration obtained from a mixture

ABBREVIATIONS

AR	Attainable region
BTX	Benzene–toluene–xylene system
CAS	Computer algebra system
CSTR	Continuous-flow stirred tank reactor
DSR	Differential sidestream reactor
IDEAS	Infinite Dimensional State-space
LP	Linear program or linear programming
MILP	Mixed integer linear program
MINLP	Mixed integer nonlinear program
PFR	Plug flow reactor
RCC	Recursive constant control policy

SECTION I

BASIC THEORY

1

INTRODUCTION

1.1 INTRODUCTION

This book is concerned with a field of study called *attainable regions* (ARs), which is a set of ideas intended to address a generalized problem, often encountered in chemical reactor and process design. Although the problem can become quite detailed if we allow it, the basic idea is simple to understand. This chapter serves to articulate the type of problems AR theory could help address. To gain a sense of the scientific discipline that we are interested in, many (but not all) of the problems we are concerned with can be represented by Figure 1.1.

Assume that we are supplied feed material of a known state (i.e., composition, temperature, and flow rate) and asked to design a process that optimizes the production of a desired product. The system in question is represented by a single block, a “black box.” This is a process that accepts a feed and converts it into something of higher economic or social value, chiefly via *reactive* unit operations. The block could be a batch or continuous process. In this book, we will mainly focus on continuous reactors. Products from the block are given by a single effluent stream. In reality, this could come about from a number of intricate recycles and bypass streams, occurring amongst a vast set of unit operations within the block—that is, the physical process represented by the block in Figure 1.1 is likely to be very complex in reality.

For many problems, we might specifically wish to find the best concentration of a desired component exiting the network, or perhaps the total cost of equipment to meet a minimum level of production—at a minimum product quality—within the network. For now, we shall leave these descriptions open so that we can describe the central problem that is related to them all.

A relevant question to ask in this context is the following: How do we best design the internals of the block given in Figure 1.1? Since we are specifically interested with reactive processes, how do we best design a process involving

reaction? There is no assumption of a predefined design inside the block. We are concerned with the generalized design of the internals, and not simply the optimization of a set design. (Some readers may recognize this as *synthesis*.) These questions often constitute what is referred to as the *reactor network synthesis problem*. Moreover, once we have synthesized a design, how do we measure a “good” design from other competing designs? Although it may appear as the former problem is our chief concern, it is actually the latter problem that we interested in. Our goal in this book is to understand how we find and measure “good” designs using AR theory.

AR theory is hence a set of ideas in chemical reactor design that aims to understand the reactor network synthesis problem. But to understand this problem will ultimately require us to understand a broader problem of what it means to be “the best,” for various designs within the block may achieve the same outcome. This is our primary concern in AR theory. It is related to the reactor network synthesis problem, but it is also distinct.

1.2 MOTIVATION

1.2.1 Toluene Production as a Case Study

To motivate why AR theory is useful, let us begin with a story, involving a team of three young engineers, who are interested in the production of toluene. We would like to gain a sense of some of the typical design considerations encountered by the team, which may also be addressed with AR theory. Sections 1.2.2–1.2.5 describe the team’s story, whereas Section 1.2.6 explains the story in relation to AR theory. Sections 1.3 and 1.4 are intended for readers who are already familiar with the rudiments of chemical reactor design.

Sam, Alex, and Donald are promising young engineers working for a large chemical company that produces toluene ($C_6H_5CH_3$) as one of its products. One day, their boss asks

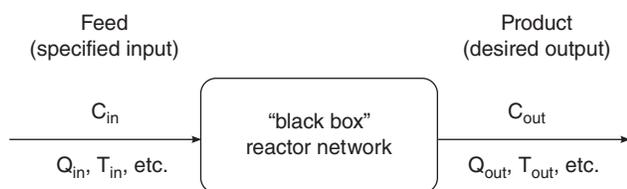


Figure 1.1 Overview of reactor network.

them to investigate how the company could improve the quality of their toluene product. Specifically, he asks:

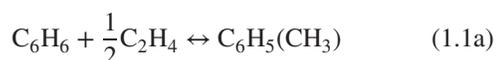
How can we maximize the amount of toluene produced from a feed of 0.5 moles of ethylene and 1.0 moles of benzene?

Although there are potentially many unit operations that might improve the amount of toluene in the system, we are mainly interested in unit operations that involve reaction. The objective may be refined slightly to say that we wish to find a *reactive* system that maximizes the amount of toluene produced.

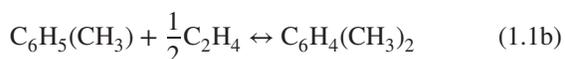
The three engineers decide to first conduct a number of experiments in the company's lab, as a way to gain a deeper understanding of the system of reactions. The discussions given in the following text chronicle their experimental process, their discoveries, and ultimately their recommendations in addressing this problem.

1.2.2 Part One: Initial Investigations

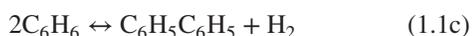
Having no prior knowledge or experience with toluene and its production, Sam first consults one of the senior chemists in the company, and gathers information regarding the reaction chemistry. The following set of reactions explain the system, including all significant products and by-products.



Benzene + $\frac{1}{2}$ ethylene \leftrightarrow toluene



Toluene + $\frac{1}{2}$ ethylene \leftrightarrow xylene



2 benzene \leftrightarrow diphenyl + hydrogen

Benzene (B) reacts with ethylene (E) to form toluene (T). Toluene and ethylene further react to form xylene (X). At the same time, benzene reacts to form two side-products in a competing side reaction: diphenyl (D) and hydrogen (H). This system may be classified as a series-parallel reaction, with toluene as an intermediate product, which shall hereafter be referred to as the BTX system.

TABLE 1.1 Sam's First Few Experiments

Experiment	Reaction Time (h)	Toluene Concentration (mol/L)
1	5.0	0.0087
2	2.1	0.0373
3	1.1	0.0560
4	0.1	0.0361

For the purposes of the investigation, the BTX reaction is independent of both temperature and pressure. It is assumed that the toluene concentration inside the reactor is simple to measure, so that concentrations are easily recorded, and that it is also possible to initiate and terminate the reaction easily.

Sam then begins her investigation in the simplest manner possible: by experimenting in a batch reactor (a beaker) in the lab, using an initial feed concentration of 0.5 mol/L ethylene and 1.0 mol/L benzene.

She starts by carrying out the experiment for a number of (arbitrary) reaction times using the same initial conditions each time, and then by recording the associated toluene concentration achieved after each run. Sam conducts four experiments in total, and a summary of her results is displayed in Table 1.1.

From the data, there appears to be an ideal reaction time, between 0.1 and 2.1 h, where the toluene concentration is maximized. Using this insight, Sam carries out a series of additional experiments, attempting to isolate the exact reaction time that maximizes toluene concentration. In effect, her approach is to use reaction time as a parameter to find the maximum toluene concentration.

After many experiments, Sam finds that a reaction time of 35.4 min (0.59 h) produces the highest toluene concentration: 0.0619 mol/L. Confident in her results, Sam reports back to her boss with the good news.



OBSERVATION: Initial investigations

Many of us tend to begin our investigations in a similar manner to Sam's approach—with little previous knowledge or known insight, we simply begin with that *is* known and then attempt to improve the current performance of the system using known parameters. This is a reasonable approach, but additional insight into the system might bring about further improvements.

1.2.3 Part Two: Iterative Improvement

1.2.3.1 Accidental Improvement Happy with Sam's initial results, the team decides to run the reaction on a larger scale. The reaction is predictable and simple to operate, and they manage to consistently achieve the toluene

concentration expected with little complications. A standard operating regime is eventually developed, involving four steps: filling, reacting, emptying, and cleaning.

One day, whilst preparing the reactor for the following day, Alex forgets to empty the entire contents of the reactor vessel, and a small portion of the reactor product is retained in the reactor overnight. Alex enters the lab the next day, ready to complete a new set of batches, unaware of his mistake the previous day. He fills the reactor with fresh feed material, and runs a new batch.

To his surprise, when he expects to obtain the same toluene concentrations as in the previous experiments, he realizes that the toluene concentration has changed. It has *increased*.

Indeed, his absentmindedness has turned out to be something of great interest—not only is it possible to adjust the reaction time of a particular batch, but it is also possible to adjust the starting concentration of the batch, using product from a previous run. With this vision, Alex proposes a new three-step reaction procedure as follows:

1. First, running a fresh batch with feed material for a restricted period;
2. Then, retaining a small fraction of product;
3. Last, refilling the reaction vessel with fresh feed again, thereby altering the starting concentration of the next batch. This batch is then run to the desired exit concentration.

Alex runs the reaction with the new operating procedure (for an arbitrary reaction time and mixture concentration) and finds that he is able to achieve a toluene concentration of 0.0652 mol/L in the following batch, which is an improvement over what Sam could achieve.

1.2.3.2 Experimenting with Different Combinations

Noting Alex's achievement, Donald begins to experiment with different retained fractions, in an attempt to find further improvements. He begins with an arbitrary combination of reaction times and mixing fraction, but he finds that some

% Reactor Volume retained	Reaction time				
	0.1 hours	0.5 hours	1.0 hours	1.5 hours	2.0 hours
20%	0.0633	0.0652	0.0644	0.0630	0.0616
40%	0.0643	0.0677	0.0659	0.0630	0.0603
60%	0.0645	0.0690	0.0659	0.0612	0.0571
80%	0.0639	0.0673	0.0624	0.0559	0.0500

Figure 1.2 Donald's investigation, summarized in a matrix for the maximum toluene concentration achieved.

combinations produce *worse* results, whereas others offer moderate improvements. He writes down his results in a matrix format, which are illustrated in Figure 1.2.

The optimal combination from Donald's finding suggests that 60% of the reactor product volume must be retained and a reaction time of 0.5 h must be used. This gives a toluene concentration of 0.0690 mol/L, or approximately 11% over Sam's investigation. Happy with this result, Sam, Alex, and Donald report back to their boss with the updated operating procedure.



OBSERVATION: Refining the approach

Although Donald, Sam, and Alex have managed to improve the toluene concentration, finding improvements are becoming more complicated in general. This procedure now involves finding an optimal combination of parameters, as opposed to just one parameter with Sam's investigation.

1.2.4 Part Three: Coffee

One morning, Donald walks into the office kitchen to make a cup of coffee. He likes his coffee strong and black. He sits the cup down on the counter and accidentally brushes past a nearby jug of milk. The jug knocks over and some of the milk spills into Donald's cup, mixing with the coffee and changing the color of its contents slightly.

Donald notes the change in the color of the coffee. Rather than discarding it and pouring a new cup, he realizes that his cup is like a beaker, and the addition of milk has changed the beaker's contents. An interesting idea appears in Donald's mind—could material be added into the toluene reactor *during* reaction? He realizes that Sam and Alex have been constraining themselves to the operation of standard batches only. But the reaction could be carried out in a *fed-batch reactor*, such as in Figure 1.3.

Donald sets out to see if he can improve the toluene concentration using fed-batch reaction. He allows material to flow into the reactor and mix with the reactor contents for the entire duration of the run, recording the maximum toluene concentration achieved after each run. After many experiments, Donald believes that he has found the optimum toluene concentration: 0.0761 mol/L; obtained by feeding at a constant rate of 1.0 L/h during reaction. This result is again an improvement over previous methods.



SIDE NOTE: New options available

The introduction of a new mode of operation has opened up a wider choice of potential operating approaches to pursue.

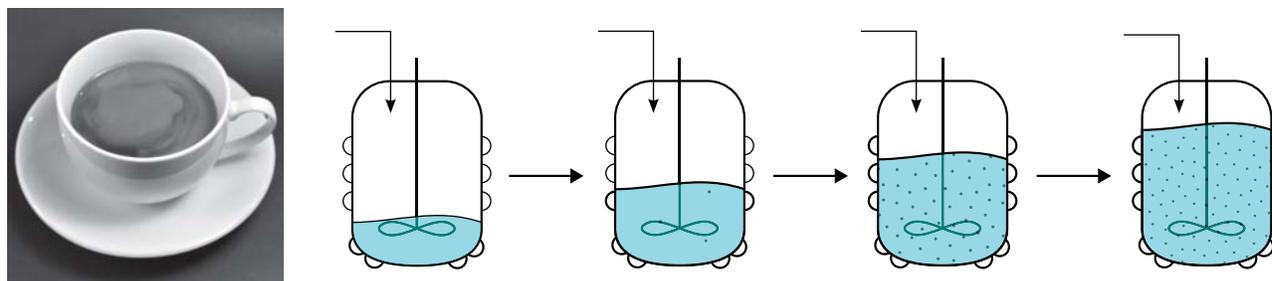


Figure 1.3 Coffee and fed-batch reactors.

1.2.5 Part Four: Additional Improvements

The team believes that they are close to reaching a point where no further improvements can be made. One day, Sam forgets to switch off the reactor just after a batch has reacted. She turns off the feed to the fed-batch reactor but forgets to turn off the reactor itself, and the reaction proceeds with a feeding rate of zero.

To her surprise, an even higher toluene concentration is obtained. Sam has, in effect, created an operating *sequence* involving the following two distinct reaction steps:

1. Reaction period involving fed-batch operation;
2. Reaction period of standard batch operation with no additional feeding.

The team discovers that if the fed-batch portion of the sequence is allowed to run for 1.6 h with a feeding rate of 1.0 L/h, followed by period of no feeding for another 15.6 min (0.26 h) as a standard batch, then a maximum toluene concentration of 0.0793 mol/L may be achieved, which again is an improvement. A summary of all the team's discoveries and recommendations for this investigation is provided in Table 1.2.

The team has managed to improve the toluene concentration by 28%: starting from 0.0619 mol/L, initially obtained by a standard batch reactor, to 0.0793 mol/L, obtained by a combined fed-batch and batch sequence. Satisfied with their progress—as well as a little tired of having to do so many

experiments—Sam, Alex, and Donald report their findings as final recommendations.



SIDE NOTE: Additional modifications

Note that concepts from Section 1.2.4 could also be incorporated into this operating sequence. Depending on how long we react, and how much product is mixed, the result might be improved even further. Each time a new discovery is found, many more combinations with existing procedures can be explored.

1.2.5.1 A Change in Objective Sam, Alex, and Donald's boss is so impressed with their findings; he proposes another problem for them to tackle (one that the company has not yet been able to fully understand): instead of seeking to maximize toluene concentration, what can be done to minimize the production of hydrogen as a by-product?

Although the team has developed useful insights from their toluene experiments, a new set of experiments must now be carried out for hydrogen, and they have little understanding that could elicit an immediate recommendation for this new objective. Their boss' request leaves them feeling a bit bewildered, and they walk away hoping that they will be as fortunate with their new investigation as they were with the previous experiments.

TABLE 1.2 All Discoveries and Recommendations Made by Sam, Alex, and Donald

	Toluene Concentration (mol/L)	Associated Method Used
Part One	0.0619	Standard batch reactor. Optimized for reaction time.
Part Two	0.0690	Repeated batch with partial mixing. Optimized for reaction time and starting concentration.
Part Three	0.0761	Fed-batch reactor at 1.0 L/h. Optimized for feeding rate.
Part Four	0.0793	Fed-batch for 1.6 h at 1.0 L/h, followed by a standard batch optimized for 15.6 min (0.26 h).



ILLUSTRATION: Results from a number of random experiments

When the objective of the investigation is modified, a new set of experiments must be carried out. Perhaps the team can use what they have learnt previously for the new investigation, but it is unclear whether the same techniques will also work with hydrogen.

Suppose that a new set of experiments are carried out, in an attempt to understand the hydrogen minimization problem, and that Table 1.3 provides a summary of those experiments. These data originate from the same BTX reaction used in the previous sections.

Which experiments would you recommend for further investigation? How much more improvement do you think is possible?

TABLE 1.3 Experiments for Minimum Hydrogen Production

Attempt	Operating Method	Hydrogen Concentration (mol/L)	Toluene Concentration (mol/L)
1	Batch reactor (0.3 h)	0.4042	0.0563
2	Batch reactor (1.5 h)	0.4396	0.0484
3	Fed-batch reactor with 0.1 L/h, 5 h	0.3682	0.0641
4	Fed-batch reactor with 1.0 L/h, 5 h	0.3677	0.0676
5	Batch reactor (0.55 h), followed by a fed-batch (0.7 h, 0.2 L/h)	0.3621	0.0685
6	Standard batch (0.5 h) mixed with 15% fresh feed at the end	0.3601	0.0522

1.2.6 What this Book is About

1.2.6.1 Attainability The challenges faced by Sam, Alex, and Donald might also be commonly encountered by those who work in a design and experimental environment. Not only is it important for Sam, Alex, and Donald to understand how to improve the reaction, but it is also important for them to understand *what* procedure should be improved and *why* it is needed. Had they been aware of this knowledge from the start, they may have arrived at their final recommendation much quicker and been more confident in their recommendations overall.

What is known in the present moment influences our thinking—in terms of what improvements are currently possible—and how these improvements should be carried out. We may be accustomed to a certain way of thinking, and sometimes our decisions depend on events that were not originally planned. Hindsight is often hard-earned. Furthermore, sometimes we do know what design or procedure should be followed, but they cannot be implemented due to financial or physical constraints. Do we always understand what impact these constraints have in terms of a potential loss, an opportunity cost?

Could we have foreseen these challenges from the beginning? How do we find these limits? And what tasks must be done in order to achieve them? There is a theme of *attainability* that runs through these questions, and AR theory provides

a framework for helping to address these questions in chemical reactor networks.

1.2.6.2 Performance Targets AR theory is concerned with problems related to the attainability of certain states. Understanding what states are attainable and what states are not allows for *performance targets* to be established, which may be incorporated into the design process. Examples of common performance targets in reactor design might include the following:

- Minimizing the production of CO₂ or other unwanted by-products from the reactor;
- Finding the smallest reactor volume needed to meet a desired output;
- Achieving the highest production rate of a biological product from a batch reactor;
- Maximizing the profit from the sale of a reactor product;
- Determining the maximum operating temperature within the reactor.

AR theory also helps us to understand what equipment is needed to achieve these targets. In relation to reactive equipment, this means providing insight into what type of reactor should be used, and how different reactor types should be

arranged to achieve a certain target, such as concentration, conversion, yield, and profit.



SIDE NOTE: Optimal result still unknown

We have not yet confirmed whether a toluene concentration of 0.0793 mol/L, achieved in Section 1.2.5, is the highest concentration possible. In Chapter 7, we shall solve this problem again and show that a toluene concentration of 0.0807 mol/L is achievable. Moreover, we can also answer the hydrogen minimization problem, described in Section 1.2.5.1, using previous calculations involving the toluene maximization problem. Hence, both questions may be addressed without the need for additional work to be carried out.

1.3 REACTOR NETWORK SYNTHESIS

In Section 1.2, we described how Sam, Alex, and Donald approached the BTX problem from an experimental perspective. How might our approach change if we are given mathematical expressions for the rates of reaction? In the following sections, we wish to describe some common ideas and approaches in theoretically designing a network of reactors (the reactor network synthesis problem), and also describe a central challenge faced in reactor network synthesis, even when mathematical and optimization techniques are available. For example, suppose that kinetics is also available for the BTX reaction and assumed to follow the data in Table 1.4:

TABLE 1.4 Rate Expressions for the BTX System

Component	Rate Expression
Benzene (r_B)	$-k_1 c_B c_E^{0.5} - 2k_3 c_B^2$
Ethylene (r_E)	$-0.5k_1 c_B c_E^{0.5} - 0.5k_2 c_T c_E^{0.5}$
Toluene (r_T)	$k_1 c_B c_E^{0.5} - k_2 c_T c_E^{0.5}$
Xylene (r_X)	$k_2 c_T c_E^{0.5}$
Diphenyl (r_D) = Hydrogen (r_H)	$k_3 c_B^2$

where $k = 1.0 \text{ L}^{0.5}/(\text{mol}^{0.5} \cdot \text{h})$, $k_2 = 1.0 \text{ L}^{0.5}/(\text{mol}^{0.5} \cdot \text{h})$, and $k_3 = 10.0 \text{ L}/(\text{mol} \cdot \text{h})$; c_i represents the concentration of component i in the system. Since expressions for species reaction rates are known, it is possible to model the BTX reaction, and hence mathematical optimizations may be carried out.

Perhaps, as a first attempt, the BTX reaction could be compared using a number of different, common, reactor models, such a continuous-flow stirred tank reactor (CSTR). One could solve the CSTR equation and then compare the

toluene concentration achieved in a CSTR to that achieved in a standard batch reactor (or equivalently in a plug flow reactor or PFR). Figure 1.4 illustrates the results of this approach using the BTX kinetics given in Table 1.4. Different reactor types behave uniquely, and thus each reactor type could be separately optimized for maximum toluene production.



SIDE NOTE: Reactor equations

If the density is assumed to be constant, a steady-state mass balance over a CSTR gives the following expression¹:

$$Q(c_i^{\text{out}} - c_i^{\text{in}}) = V_{\text{CSTR}} r_i \quad (1.2a)$$

Here, c_i^{in} and c_i^{out} are the inlet and exit concentrations of component i , respectively. Variables Q and V_{CSTR} are the volumetric flow rate and CSTR vessel volume, respectively. Similarly, the equation for a constant volume batch reactor follows the differential equation:

$$\frac{dc_i}{dt} = r_i \quad (1.2b)$$

where t is the reaction time of the batch. Solving Equation 1.2a and 1.2b produces the toluene concentration profiles in Figure 1.4.

In this case, the CSTR achieves a higher toluene concentration, but are there perhaps other reactor types (i.e., a packed bed reactor) that could outperform the CSTR? Additionally, are there *combinations* of reactors—as a *network of reactors*—that could achieve better?

To demonstrate the potential complexity involved when dealing with multiple reactor designs, consider Figure 1.5, which proposes a number of different reactor configurations (reactor structures) that might be used. For simplicity, the configurations are limited to combinations of a maximum of three reactors, using PFRs and CSTRs only.

How do we determine which reactor configuration is best? In this example, we could potentially solve each system and find the best configuration from the size options. However, we can always devise new configurations that may be superior. A number of additional design choices must be made when considering reactor structures, even for simple configurations such as in Figure 1.5. Some of these considerations might include the following:

- How many individual reactors do we consider in each structure?
- What reactor types do we consider?

¹A summary of fundamental reactor types is provided in Appendix A.

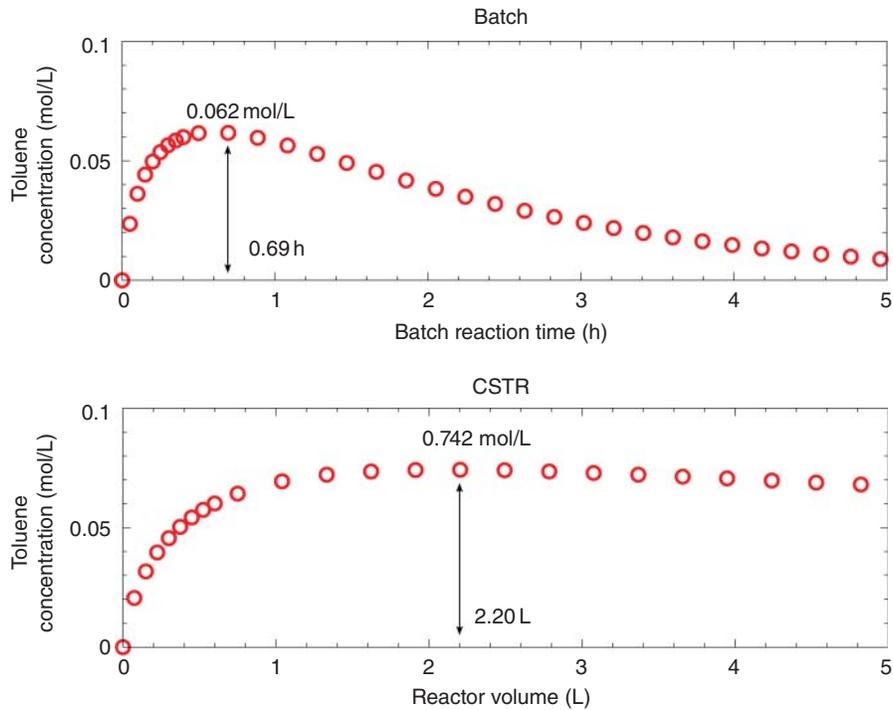


Figure 1.4 Toluene concentration data obtained in a batch experiment and a continuous-flow stirred tank reactor (CSTR).

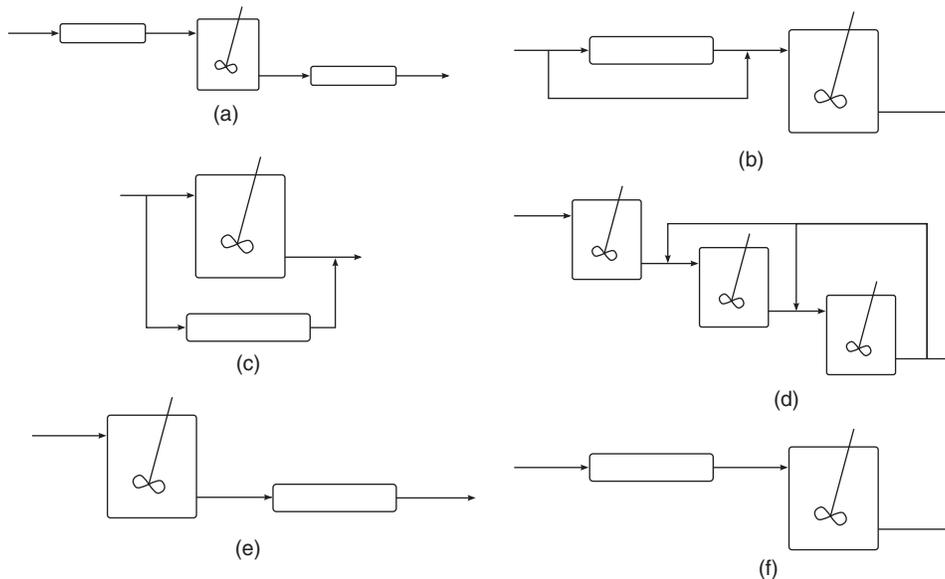


Figure 1.5 Options (a)–(f) represent a number of different reactor configurations. Only PFRs and CSTRs are used with a maximum of three reactors per configuration.

- Do we include recycle and bypass streams? If so, where are they placed within the structure?
- How many parallel reactors are included?
- How many reactor structures should be considered?

How should one select the best reactor structure from a pool of many reactor configurations? Nominating specific combinations in an exhaustive fashion is not feasible because we could always devise a different configuration that may perform better. Yet again, we are presented with a

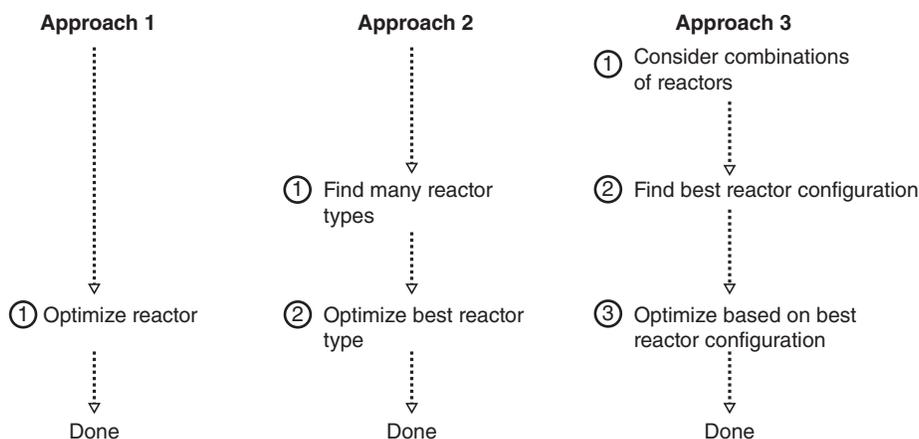


Figure 1.6 Overview of different approaches. Each approach will terminate in an optimal design, although the particular design achieved may differ depending on the order of steps taken.

similar question as in Section 1.2.6—*when do we know we are the best?*



CONCEPT: Reactor structures

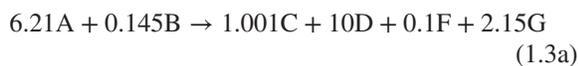
By a *reactor structure*, we have in mind a well-defined arrangement of reactors (a reactor network) that produces a particular output. The network might typically be composed of various reactor types. The kinds of problems that we will investigate in this book all fall into this category. We will be interested in looking at how combinations of reactors (as opposed to a single reactor) can often provide meaningful improvements to a problem.

Figure 1.6 proposes a number of general approaches to the reactor synthesis problem, starting from the simplest and most constrained approach, to the most general (and difficult) approach. In AR theory, we are ultimately concerned with problems involving reactor structures that produce the best performance, and thus AR theory falls into Approach 3 of Figure 1.6.

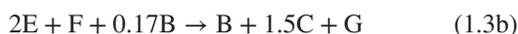
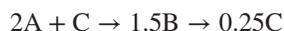


ILLUSTRATION: Are multiple reactors necessary?

Consider the following two reactions:



and



How would you build a reactor that maximizes the production of component C according to Equation 1.3a? How would you optimize for the same scenario if Equation 1.3b occurred in the reactor instead?

In Equation 1.3a, the reaction is “simple”: although multiple components participate in the reaction, there is only one reaction, and hence there is only one pathway that reactants may proceed to form products (all components are linked by a common extent of reaction). The choice of reactor type *may* have a significant influence on system performance, although it is possible to achieve the same performance with different reactor types.

By comparison, Equation 1.3b contains the same number of individual components, yet these are split over a number of different reaction paths. Equation 1.3b is hence more complex: each path could be individually pursued, resulting in a number of different product mixtures depending on which reactions are dominant, the initial concentration of species, and the intrinsic behavior of different reactor types to these reactions. Consequently, the choice of reactor type may have a significant influence on performance. One reactor type may only achieve a small portion of all possible product states, favoring one reaction over other competing reactions—a CSTR might favor the first two reactions, whereas a PFR might favor the last reaction. Utilizing different reactor types may expose a larger set of possible outputs, but the opportunity for different reactions to occur means that the underlying reactors must be sufficiently generalized to target the operating point of interest. In order to achieve these states, reactor structures are more suitable.

ILLUSTRATION: Biological reactions

Consider the metabolic pathway diagram in Figure 1.7 for the production of nattokinase (and other biological products) by glucose and glycerol. Here, there are *many* reactions occurring. Each arrow displayed in the schematic represents a reaction.

The diagram given in Figure 1.7 is common in biological systems. Complex recycling of intermediate products, and the possibility for compounds to traverse along many divergent paths, means that it is often not straightforward to predict how the flow of material is best arranged along a certain pathway, or what the best pathway is for the production of a desirable intermediate product. Modeling the reactions present in a biological system is complex, and optimizing for a bioreactor that maximizes the production of a specific byproduct is hence a challenging task.

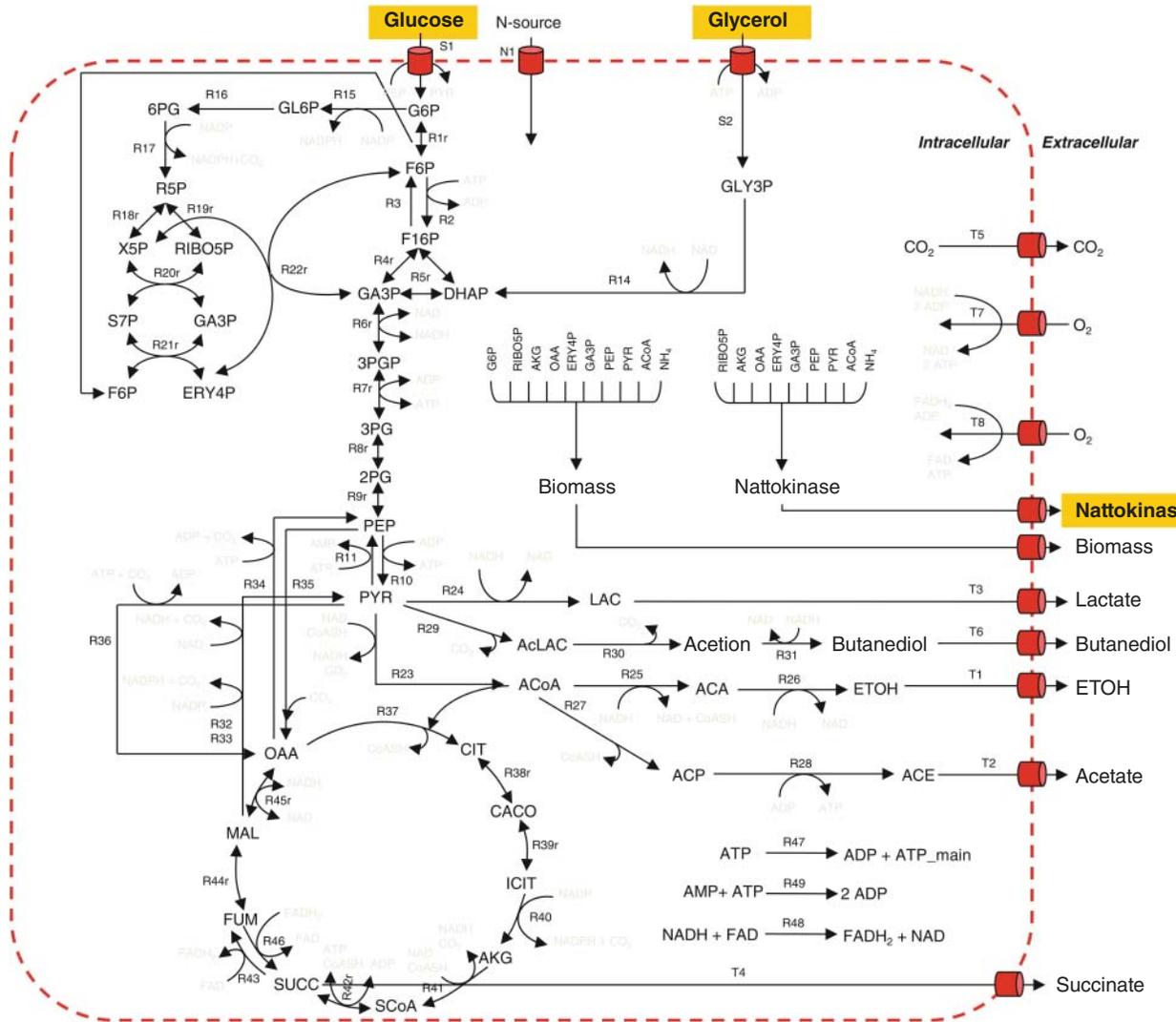


Figure 1.7 Metabolic pathway showing how glucose and glycerol are converted in a series of complex biological reactions to produce a number of different by-products. Unrean and Nguyen (2013). Reproduced with permission of Springer.

There is again an opportunity here to use multiple reactor types to maximize a desired product, yet in practice, simplifications are often applied (i.e., lumped reaction models) to make the problems more tractable. The design of the reactors themselves are also often of a simpler nature, employing a single CSTR (chemostat) or fed-batch reactor with biomass recirculation (Nielsen et al., 2011). Utilizing one reactor in one configuration may limit the possible combination of pathways, which may limit performance as a result.

Although multiple reactors can be used for single reaction systems, they are potentially not required. For systems involving multiple reactions, reactor structures may be *essential*—unlocking certain states that would not

be possible with a single reactor type. However, solving reactor network synthesis problems is more difficult than single reactors, because reactor structures introduce more complexity into the design.



SIDE NOTE: Multiple reactions and multiple reactors

We find that methods such as AR theory are required not because of problems involving multiple reactors, but because of problems involving multiple reactions. Many of these methods are unnecessary when the reactions are inherently simple. The reactor network synthesis problem arises often as a result of complexities in the system from multiple reactions.

When the reaction is complex, the best performance is often achieved in a reactor network (a combination of reactors). AR theory deals with reactor problems involving *more than one reactor*.

1.4 SOLVING THE REACTOR NETWORK SYNTHESIS PROBLEM

1.4.1 Reactor Superstructures

1.4.1.1 Description A popular approach to solving the reactor network synthesis problem is by use of *reactor superstructures*. A reactor superstructure is a reactor configuration

where many simpler reactor types are arranged in a specific pattern and connected via a network of bypass and recycle streams, so that various outcomes can be achieved from a single structure. Reactor superstructures thus represent a superset of all possible outputs, wherein an optimal answer is a subset of the superstructure.



ILLUSTRATION: Examples of reactor superstructures

Consider how a series of CSTRs might act as potential building blocks that approximate other reactor types.

For PFR approximation, CSTRs can be connected in series, as in Figure 1.8(a). A variety of different outputs may be produced from a single reactor type in the appropriate arrangement. More complex reactor structures have been published in the scientific literature. One example is provided in Figure 1.8(b) (Rooney and Biegler, 2000).

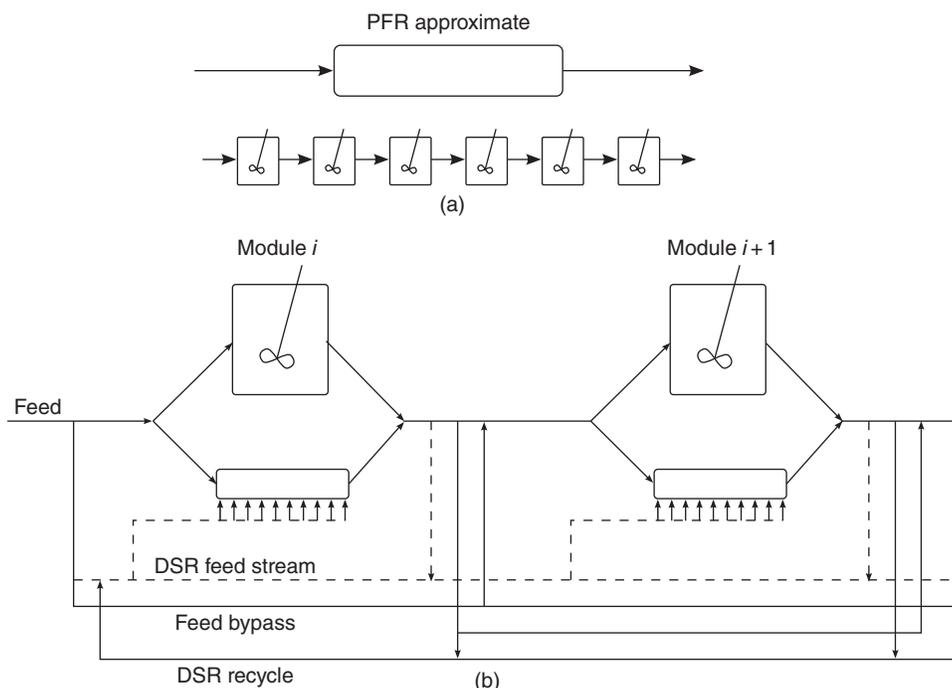


Figure 1.8 (a) A CSTR configuration that approximates a plug flow reactor (PFR). Kauchali et al. (2002). Reproduced with permission of Elsevier. (b) Example of a reactor superstructure. Rooney and Biegler (2000). Reproduced with permission of Elsevier.

Combinations of basic reactor types, such as CSTRs and differential sidestream reactors (DSRs), are arranged in a specific pattern to produce a building block structure for the creation of more sophisticated reactor structures. These superstructures allow for the description of a wider variety of different achievable states.

Using the superstructure approach, nonreactive unit operations (i.e., separation) may be included into the design as well, which allows for a wide range of optimization scenarios to be solved for. The superstructure approach is a powerful method to reactor network design, as a large amount of detail and flexibility can be incorporated into the configuration.



SIDE NOTE: The power of the superstructure approach

Superstructure methods are a powerful and popular approach to the reactor network synthesis problem, as well as to the generalized problem of process synthesis (the design of entire processes). An advantage of superstructures is that they interface easily with existing plants—that is, it is often possible to mathematically model the operation of plants that have already been designed and built, which once represented as a mathematical model, can be optimized with standard optimization methods. This makes the superstructure approach highly useful in industry, as existing plants can be modeled and optimized directly, making results much easier to relate to real-life performance.

1.4.1.2 Solving Optimal Reactor Superstructures With a large number of interlinked bypass and recycle streams, amongst a number of different reactor types, reactor superstructures are expressed in a large mathematical model—a system of differential algebraic equations (DAEs)—which can be solved using mathematical optimization techniques. To solve the model, the parameters of the superstructure model are adjusted and checked against an appropriate objective function to determine optimal performance. Optimization follows the generalized form:

Maximize/minimize:	Objective function (i.e., volume, concentration, time or operating costs)
Subject to:	Parameters and constraints (i.e., time, reactor model equations, flow rates and mixing fractions)

The complexity of modeling and optimizing the superstructure is then related to the complexity of the superstructure itself. Parameters in the model act like switches on a switchboard, and a solution to the optimization problem is

a specific combination of switches that achieves the desired result. A balance must be established between practicality and accuracy: the superstructure must be adequately generalized (more complex) to describe many outcomes, yet computationally simple to allow for solution of the associated problem in a reasonable time.

Solutions are based on the objective function specified. A change in the objective function introduces a change in the superstructure configuration, which might require re-solving the problem for the new objective function. (When Sam, Alex, and Donald are asked to minimize the production of hydrogen in the BTX reaction, this signals a change in objective function.)

Two important questions that arise when dealing with the optimization of reactor superstructures thus arise:

1. Are there similar superstructure configurations that achieve the same result? (*Are there multiple solutions?*)
2. Does a better superstructure exist? (*Is the answer globally optimal?*)

More generalized structures, capable of describing a wider variety of solutions, may require additional complexity in the superstructure model. Additional complexity in the design also leads to increased complexity in the solution of the equations describing the superstructure, which may make the problem more difficult to solve. (Figure 1.8(b) might be able to describe more complex problems and solutions based on Figure 1.8(a) may offer better performance than Figure 1.8(a), but it is also more difficult to solve.)



ILLUSTRATION: Word combinations

How many unique English words can you form from the following set of letters? Not all letters in the set need be used:

{C, X, A, H, T}

Suppose now that the letter “E” is included into the set. How many more words can you find? What if letters “D” and “O” are added to the set?

With the help of a computer, we can show that there are approximately 11 unique words that can be formed from the set {C, X, A, H, T}, 32 unique combinations are possible from the set {C, X, A, H, T, E}, and 83 words are possible from {C, X, A, H, T, E, D, O}. Indeed, it is evident that with each addition of a new letter, there