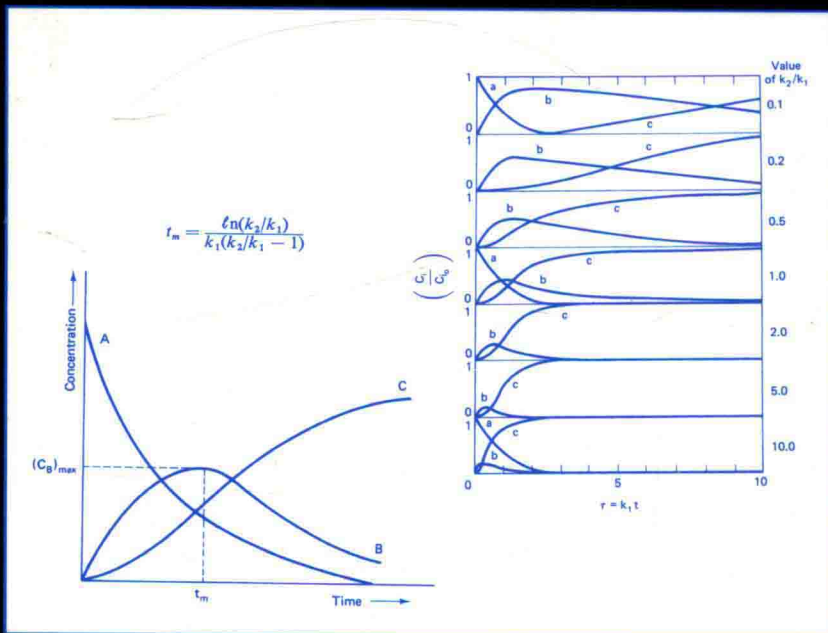


# REACTION KINETICS AND REACTOR DESIGN

*Second Edition, Revised and Expanded*



**John B. Butt**

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*Second Edition, Revised and Expanded*

**John B. Butt**

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#### **ADDITIONAL VOLUMES IN PREPARATION**

## Preface to the Second Edition

A lot of things have happened in chemical reaction engineering since the first edition of this book, and this second version is very much different from the first. The remarks here are confined mostly to pointing out the changes.

The first edition was designed to combine a thorough description of the origin and application of fundamental chemical kinetics all the way through to realistic reactor design problems. This is continued, although the present effort increases the applications to a much larger range of reactor design problems. The discussion of kinetics has been expanded in Chapter 1 to include material on chemical thermodynamics related to reaction systems and additional discussion of chain and polymerization reactions. New material on microbial and enzyme kinetics, and on adsorption-desorption theory, is given in Chapter 3. At the same time, some of the more deadly derivations arising from the kinetic theory of gases given in the original Chapter 2 have been streamlined and the presentation simplified in the present version. Hopefully this material, which is very important to the philosophy of this book, will now be more digestible.

Chapters 4–6 retain the general organization of the first edition, although the presentation has been expanded and brought up to date. After the first part of Chapter 7 on transport effects in catalytic reactions, the present book goes its own way. A discussion of gas–solid noncatalytic reactions is given, and the development of two-phase reactor theory based on plug flow, mixing cell, and dispersion models (including transport effects and nonisothermality) rounds out the presentation of the chapter. This then leads naturally into the detailed multiphase reactor design/analysis considerations of Chapter 8, which treats some specific types of multiphase reactors such as fluid beds and trickle beds. This material is generally oriented toward design considerations, but care has been taken to relate the presentation to the pertinent theoretical developments given in earlier chapters.

Finally, Chapter 9 deals with unsteady-state fixed-bed problems, including catalyst deactivation phenomena, adsorption, ion-exchange and chromatographic reactors. This chapter probably has less to do with chemical reactions per se than the others, but the foundations of the fixed-bed analyses employed are firmly established in the prior material, and it seems a waste not to make use of it here.



So much for the menu. The first edition was designed primarily as a text for an undergraduate course. This edition, as one might expect from the above, goes considerably beyond that. According to the desires of the instructor, portions can be used for undergraduate courses, other parts for graduate courses, or special topics selected for seminar or discussion purposes.



HORATIO SAYS

There are a number of major changes, in addition to the extent of coverage, to be found here. A major new feature is the inclusion of a large number of worked-out illustrations in the text as the narrative continues. These can be fairly comprehensive and are not always easy; hopefully they are indicative of what is to be expected in the "Exercises" at the end of each chapter. We also have a house pet mouse, "Horatio," who appears at the end of almost all of the illustrations, asking further questions regarding the illustration—essentially giving another set of exercises at the choice of the instructor. They are effective in providing additional experience with specific topics as desired. Horatio also appears in several locations asking the reader to clarify for himself various points in the text. Thanks to Mr. David Wright for permission to use this figure.

Many references are given in the text, as we go along, to pertinent background material. It has been an objective here to go back to sources that presented the early, developmental work on a given topic, and to supplement those references with later ones presenting significant advances. A consequence of this is that one should not be surprised to see references from the 1950s, or even much earlier, from time to time. Another procedure that has been followed is to use the notation as generally employed in the source material to make it easier in reading those references. As a result, there is no set of standard notation in the book; however, all the notation used in each chapter is summarized as completely as possible, alphabetically, at the end of that chapter.

Computer literacy is assumed, and there are many problems that require computer solution, particularly as one becomes involved with nonisothermal reactors, boundary-value dispersion problems, the more advanced fixed-bed problems, and interpretation of kinetic data. We have not tried to get into the software business here, in view of the continuing rapid evolution of various aspects of that field. We have yielded to the temptation in a couple of instances to suggest, in outline, some algorithms for specific problems, but in general this is left up to the reader.

The indebtedness to colleagues, teachers, and students mentioned in the preface to the first edition remains unchanged. In addition, I would like to thank Professor James J. Carberry and Professor Jon H. Olson for their comments on the preparation of this second edition.

*John B. Butt*

## Preface to the First Edition

All I know is just what I read in the papers

— *Will Rogers*

It is probably obvious even to the beginning student that much of chemical engineering is centered on problems involving chemical transformation, that is, chemical reaction. It is probably not so obvious, at least in the beginning, that the rate at which such transformations occur is the determining factor in a great number of the processes that have been developed over the years to produce that vast array of goods that we consider an integral part of contemporary life. The study, analysis, and interpretation of the rates of chemical reactions is, itself, a legitimate field of endeavor. It ranges in scope from those problems concerned with the fundamentals of detailed mechanisms of chemical transformation and the associated rates to problems that arise during the development and implementation of procedures for chemical reactor and process design on a large scale. If we must give names to these two extremes, we might call the first "chemical kinetics," and the second "chemical reaction engineering."

In the following we shall range from one limit to the other, although our primary objective is the understanding of kinetic principles and their application to engineering problems. What will we find? For one thing, we will find that chemical reactions are not simple things; those fine, balanced equations which everyone has used in solving stoichiometry problems ordinarily represent only the sum of many individual steps. We will find that the rates of chemical transformation, particularly in engineering application, are often affected by rates of other processes, such as the transport of heat or mass, and cannot be isolated from the physical environment. We will find that the normal dependence of reaction rate on temperature is one of the most intractable of nonlinearities in nature, providing at the same time many of the difficulties and many of the challenges in the analysis of chemical rates. We will find that often it is not the absolute rate of a single reaction but the relative rates of two or more reactions that will be important in determining a design. We will find that space as well as time plays an important role in reaction engineering, and in the treatment of such problems it will be necessary to develop some facility in the use of rational mathematical models. Finally, we will find that the artful compromise is as important, if not more so, in our applications of reaction kinetics as it is in all the other areas of chemical engineering practice.

This may seem like a very short list of what is to be found if the topic is truly as important as we have indicated. It is intentionally short, because the essence should not be submerged in detail quite so soon, or, to paraphrase Thomas à Kempis, ... it is better not to speak a word at all than to speak more words than we should.

The material of this text is intended primarily to provide instruction at the undergraduate level in both chemical kinetics and reactor design. Of particular concern has been the detailing of reaction kinetics beyond phenomenological description. The rationale for the Arrhenius equation was a personal mystery to the author in earlier years, who hopes an appropriate solution is revealed in Chapter 2.

Numerous other aspects of classical theories of chemical kinetics are assembled in Chapter 2 and Chapter 3 to give some perception of the origin of phenomenological rate laws and an understanding of the differing types of elementary reaction steps. In Chapter 4 we swap the beret of the theoretician for the hard hat of the engineer, in pursuit of means for developing rational chemical reactor design and analysis models. A parallelism between mixing models and reactor models has been maintained in order to demonstrate clearly how reaction kinetic laws fit into reactor design. Chapters 4 through 6 are based on homogeneous models, and proceed from standard plug flow and stirred tank analysis to description of nonideal behavior via dispersion, segregated flow, mixing cell and combined model approaches. Phenomena associated with reaction in more than one phase are treated in Chapter 7 but no attempt is made to develop multiphase reactor models. The fact that reaction selectivity as well as reaction rate is an important and often determining factor in chemical reaction or reactor analysis is kept before the eyes of the reader throughout the text.

The exercises are an intentionally well-mixed bag. They range from simple applications of equations and concepts developed in the text to relatively open-ended situations that may require arbitrary judgement and, in some instances, have no unique answer. The units employed are equally well-mixed. Historically, multiple systems of measure have been a curse of the engineering profession and such is the case here particularly, where we range from the scientific purity of Planck's constant to the ultimate practicality of a barrel of oil. The SI system will eventually provide standardization, it is to be hoped, but this is not a short-term proposition. Because both author and reader must continue to cope with diverse sets of units, no attempt at standardization has been made here.

Symbols are listed in alphabetical order by the section of the chapter in which they appear. Only symbols which have not been previously listed or which are used in a different sense from previous listings are included for each section. Symbols used in equations for simplification of the form are generally defined immediately thereafter and are not listed in the Notation section found at the end of each chapter.

Each chapter is divided into more or less self-contained modules dealing with a unified concept or a group of related concepts. Similarly, the exercises and notation are keyed to the individual modules, so that a variety of possibilities exist for pursuit of the material presented.

Acknowledgment must be made to teachers and colleagues who, over the years, have had influence in what is to be found in this text. I am grateful to the late Charles E. Littlejohn and R. Harding Bliss, to Professors C. A. Walker, H. M. Hulburt, and R. L. Burwell, Jr., and especially to Professor C. O. Bennett, who offered many constructive and undoubtedly kind comments during preparation of this manuscript.

Thanks also to R. Mendelsohn, D. Casleberry and J. Pherson for typing various sections of the manuscript, and to the Northwestern chemical engineering students for detecting unworkable problems, inconsistent equations, and all the other gremlins waiting to smite the unwary author.

*John B. Butt*

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## Apparent Reaction Kinetics in Homogeneous Systems

Gather ye rosebuds while ye may  
Old time is still a-flying

— Robert Herrick

### 1.1 Mass Conservation and Chemical Reaction

Certainly the most fundamental of laws governing the chemical transformations and separations with which chemical reaction engineering is involved is that of conservation of mass. Although this is surely not new to the readers of this text, it is worth the time here to revisit a simple example to make clear what specific functions in a typical mass balance might arise as a result of chemical reaction. The example may seem very elementary, but it is important that we all start at the same point. Consider then the steady-state separation process depicted in Figure 1.1. A stream,  $L$ , mass/time, containing two components, A and B, is fed to the process, which divides it into two product streams,  $V$  and  $W$ , mass/time, also containing components A and B. The mass fractions of components A and B in  $L$ ,  $V$ , and  $W$  are given as  $x_A$ ,  $x_B$ ,  $y_A$ ,  $y_B$ , and  $z_A$ ,  $z_B$ , respectively. Mass is conserved in this separation; we may express this mathematically with the following simple relations:

$$L = V + W \quad (1-1)$$

$$Lx_A = Vy_A + Wz_A \quad (1-2)$$

$$Lx_B = Vy_B + Wz_B \quad (1-3)$$

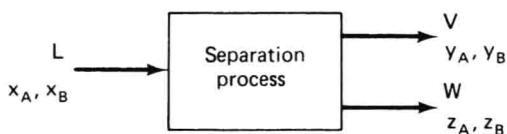


Figure 1.1 Simple separation.



Now, since we also know that each stream consists of the sum of its parts, then

$$x_A + x_B = 1 \quad (1-4)$$

$$y_A + y_B = 1 \quad (1-5)$$

$$z_A + z_B = 1 \quad (1-6)$$

This, in turn, means that only two of the three mass balance relationships (1-1) to (1-3) are independent and can be used to express the law of conservation of mass for the separation. We are then left with a system of five equations and nine potential unknowns such that if any four are specified, the remaining five may be determined. Of course, all we have done is to say:

$$\text{Total mass in/time} = \text{total mass out/time}$$

$$\text{Mass A in/time} = \text{mass A out/time}$$

$$\text{Mass B in/time} = \text{mass B out/time}$$

Since the uniform time dimension divides out of each term of these equations, our result is the direct mass conservation law.

Now let us consider a slightly different situation in which the process involved is not a separation but a chemical transformation. In fact, we shall simplify the situation to a single input and output stream as in Figure 1.2 with the feed stream consisting of component A alone. However, within the process a chemical reaction occurs in which B is formed by the reaction  $A \rightarrow B$ . If the reaction is completed within the process, all the A reacts to form B and mass conservation requires that the mass of B produced equal the mass of A reacted. The material balance is trivial:

$$L = W \quad (x_A = 1, z_B = 1) \quad (1-7)$$

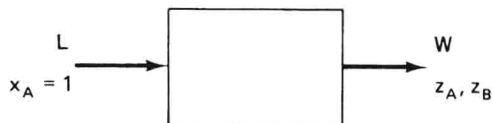
What happens, though, if not all of the A reacts to form B in the process? Then, obviously, the mass of B leaving is not equal to the mass of A entering, but rather

$$L = Wz_A + Wz_B \quad (1-8)$$

Comparison of the two processes illustrates in a simple but direct way the general concerns of this whole text. These are

1. To determine  $z_A$  and  $z_B$  given a certain type and size of reaction process
2. To determine the type and size of reaction process needed to produce a specified  $z_A$  and  $z_B$

Two factors enter into this problem. The first is the *stoichiometry* of the reaction transforming A to B. Chemical equations as normally written express the



**Figure 1.2** Chemical reaction process.