MULTIPHASE CHEMICAL REACTORS

THEORY, DESIGN, SCALE–UP

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Theory, Design, Scale-up

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Preface

Satterfield, writing in 1975, expressed surprise at the small amount of information on design, theory, and performance of trickle bed reactors published in the literature when measured against the widespread use of this reactor type by the refining and biochemical industries. His comment would have been equally valid for all types of multiphase reactors. Now, in the early 1980s the situation has changed. There has been a heavy flow of experimental and theoretical studies of this reactor class in the last decade. Examine the contents of research journals and you will see as many papers on three phase reactors as on fluidized bed or fixed bed, gas phase reactors. Important research groups specializing in three-phase reactors can now be found in universities and government research institutes around the world. Strong specialist groups exist in many of the major refining companies, although they remain tight-lipped about their research activities.

Growth of the literature in the last few years has spawned not only this monograph but also three others: L'Homme's *Chemical Engineering of Gas-Liquid–Solid Catalyst Reactions,* Shah's outstanding book *Gas-Liquid-Solid Reactor Design,* and Ramachandran and Chaudhari's *Three-Phase Catalytic Reactors.* It is no longer possible to dispose of multiphase reactors in one or two chapters of a general treatment of reactor design and do justice to the subject.

Perspective

Multiphase reactors—along with smelters and kilns—are the earliest types of chemical reactors in our civilization. Egyptian hieroglyphics dating from 2500 B.C. describe wine making; Mesopotamian records for this activity are even older. From these early records we can deduce that the importance of mixing in operating these primitive fermentation reactors was clearly understood. The need to control reaction extent and the means thereof were also known.

Waste treatment provides many more modern examples of the use of multiphase reactors, but these uses predate the emergence of reactor engineering as a distinct discipline. Activated sludge and trickling filter systems for domestic waste treatment were in use by the beginning of the twentieth century. In the chemical industry an early application (1920+) of trickle beds was the synthesis of butadiene from acetylene via a four-step process. One of the steps, hydrogenation at 300 atm, was performed in a trickle bed. In the 1940s the Reppe process for butadiene from acetylene was commercialized in Germany. The key step—the reaction of acetylene and formaldehyde at 100°C and 5 atm—over a copper acetylide to form butynediol was carried out in a trickle bed. Hydrogenation of butynediol, a further step, was performed in the same type of unit. Hydrogenation of vegetable fats in slurry reactors has been a commercial process in the food industry since about 1910. Extensively modified and using better catalysts, it is still in use today.

Truly large-scale use of multiphase reactors dates from the 1950s with their application to hydrotreating of lubricating oils, followed 5 to 10 years later by the widespread adoption of hydrodesulfurization and the installation of the first hydrocracking units.

The history of research into the operation and design of multiphase reactors is difficult to trace. Apparently, the effort has developed from two disparate directions: (1) study of gas-liquid mass and heat transfer primarily directed at absorbers and strippers and (2) single-phase reactor design. Much of the data for reactor hydrodynamics and even external mass transfer have been generated in either nonreacting systems or, if reacting, in noncatalytic ones. Treatment of diffusion and reaction in porous solids is drawn from the literature on the heterogeneous catalysis of gas-phase reactions.

Papers giving results of experiments in slurry reactors, or indeed in other multiphase reactor types, but without any analysis of the complex three phase hydrodynamic and transport phenomena, date, of course, back to the early years of the applied or industrial chemistry literature. One of the early trickle bed reactor studies, on the liquid phase oxidation of ethanol to acetic acid over a Pd catalyst, which examined the hydrodynamics specifically, dates from 1955 (Klassen and Kirk). The study of bubble column reactors for the Fischer-Tropsch synthesis was described by Kolbel, Ackermann, and Engelhardt in *Erdole und Kohle* in 1956. Research on slurry reactors and specifically biological application also dates from the early 1950s.

The literature on multiphase reactors is now expanding rapidly. Shah, writing in the preface of his recent book *Gas-Liquid-Solid Reactor Design,* calls multiphase reactors the most widely researched topic in chemical reactor engineering.

Background

This monograph was compiled from a set of notes prepared for a short course on the theory, design, and application of multiphase reactors organized by one of us (Gianetto). This five-day course was held in October, 1982, in Kitchener, Ontario, Canada, near the University of Waterloo. The purpose of this course was to draw together the significant advances in understanding of multiphase phenomena, the improvements in modeling multiphase reactors, and the mass of experimental data. It was also our aim to make this vast new information available to those charged with development, design, or troubleshooting of this reactor class.

To develop the course, we contacted those in the reactor engineering field who have established international reputations for their research on multiphase reactors or on some closely applied area of study. Naturally, limited resources curtailed the number of lecturers who could be engaged and from where they could be drawn. Unfortunately, the cost of traveling prevented us from inviting any of the outstanding Japanese researchers in the field or anyone from India, even though the research "school" built around Sharma at Bombay has attained a substantial reputation.

The number of gifted, important researchers now active on multiphase reactor problems has become so large even in North America and Europe that our choice of lecturers fails to include many important researchers. Regrettably, a number of important research "schools," represented by Shah in Pittsburgh, Satterfield at MIT, and Dudukovic in St. Louis could not be added as lecturers.

Success of the first offering in Canada led to a second offering in Frankfurt, West Germany, in July 1983, sponsored by DECHEMA. We hope to bring the course to other locations around the world in future years. Our purpose in preparing this monograph is to make the material developed for these short courses available to those unable to participate.

Objectives

The philosophy underlying this monograph is that imaginative as well as reliable engineering requires an understanding of the physics of the system and a clear recognition that the models used are approximations. It is important to know what simplifications have been made and under what conditions they can be justified.

Many phenomena interact in multiphase reactor design and operation: chemical kinetics, hydrodynamics of fluids with free surfaces, hydrodynamics of fluids moving through permeable media, turbulence, transport, and surface phenomena. Sometimes these interactions are synergistic; at other times they are destructive. Frequently, interactions are rather weak, and one or more phenomena dominate. On the other hand, interactions can be very strong and inseparable, such as diffusion and reaction in a porous solid. It follows, then, that multiphase reactor systems are complex—much more so than their single-phase, surface-catalyzed analogs. Despite the progress made in the preceding decade, our understanding of all phases of this class of reactors remains incomplete.

The objective of this volume is to deepen the reader's appreciation of the dominant physical processes occurring in the major three-phase reactor types and to provide models (and mathematical relations) useful for data reduction, a priori design from bench-scale measurements, and scale up at all stages. Our intention is to identify the simplifications in the models and suggest, when appropriate, bounds of validity.

Another objective is to draw together experimental data, often from various sources, into a useable form for engineering purposes. A conscious objective has been to point out questions or engineering aspects for which our understanding is still incomplete.

For the most part, we avoid summaries in the individual chapters. This is because each subsection in any chapter is in itself a distillation of the literature or, at least, one or more rather intensive studies. Similarly, we have generally not made recommendations as to which technique, procedure, or correlation is to be used. This is intentional because of either the limited validity of the models and relations or because there is no compelling evidence to choose one relation over another.

Organization

Technical discussion begins with an examination of the different types of multiphase reactors; their advantages and typical applications. Chapter 1 provides an overview of physical phenomena, hydrodynamics, models, and scale-up procedures.

The next six chapters examine the problem of simultaneous diffusion and reaction. Particular attention is paid to the calculation or measurement of system and/or model parameters. Problems of scale up and the justification for mock ups are also examined.

Chapters 8 through 12 form a third unit whose main theme is downflow, cocurrent reactors, commonly known as trickle beds. Upflow, cocurrent reactors, called bubble columns, are examined in Chapter 10. Chapters 11 and 12 discuss mechanically agitated slurry reactors—the third and last type of reactor considered in the monograph.

The final unit—Chapters 16 through 18—treats application. Chapter 16 considers the important application of multiphase reactors to various types of hydrogenation processes, Chapter 17 examines recent applications to coal liquefaction, and Chapter 18 looks at biochemical applications and the special problems that arise when organisms or otherwise sensitive substances make up the solid phase.

Presentation

A difficulty of a monograph such as this, with its numerous authors, is that a uniform viewpoint and presentation are impossible to achieve. The editors have attempted to make this monograph more than a collection of review papers. We have suggested that the authors delete some of their material, expand the treatment of their topic, or include research results other than those they originally used in order to make our treatment of multiphase reactors more complete, as well as to reduce overlap between chapters. Some overlap is unavoidable and even desirable to round out a discussion or to justify assumptions or simplifications. You will find many examples of this sort of overlap in what follows. We trust the repetition is helpful.

In editing the individual chapters, we tried to find a compromise between allowing each author his own style and achieving a more or less uniform presentation in North American English. You will find the ordering of each chapter to be different, and the way material is presented varies. Some chapters, therefore, may be more difficult for you to use than others, just because of how the author has chosen to develop his topic and what background he has assumed you have. No chapter, we hope, will be found totally obtuse.

We have also attempted to avoid confusion in nomenclature. A general nomenclature following the suggestions of the Working Committee on Reactor Engineering of the European Federation of Chemical Engineering has been adopted and appears after the Acknowledgments. Even this expanded set is not adequate when details are considered in each of the individual chapters. Thus, an additional notation is given at the end of each chapter. To simplify its use, it gives all the nomenclature for the chapter, repeating some of the items given in the general nomenclature.

Corrections and Additions

It is our hope to develop new editions from time to time. We would be grateful, therefore, to have our attention drawn to research or practice we have overlooked. Of course, we welcome identification of errors that have inadvertently crept into this monograph.

Agostino Gianetto
Peter L. Silveston

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