Chemical Kinetics and Catalysis

By R. A. van Santen and J. W. Niemantsverdriet, Plenum Press, New York, 1995, 280 pp., $69.50.

I am going to comment on this book from the perspectives of three different groups of readers: beginning graduate students in catalysis, instructors who are considering it for use in a course, and catalytic researchers. The first group is probably the primary intended audience because the book evolved from a course taught at the Schuit Institute of Catalysis at the Eindhoven University of Technology in the Netherlands. At the same time, since the book is part of the *Fundamental and Applied Catalysis* series aimed at both academic and industrial researchers, it should also be of interest to the other two groups.

As a student I would enjoy reading Chapter 1 because it provides a nice perspective of catalysis through a brief discussion of its history, industrial relevance, and scientific subdisciplines. Chapter 2 presents rate equations for homogeneous and heterogeneous systems. By introducing the concept of reaction affinity, the authors do a nice job of showing the relationship and consistency between thermodynamics and kinetics in situations that are either near or far away from equilibrium. Chapter 3 is not as easy to read because in trying to survey catalysis by metals, oxides, and sulfides in 32 pages, the authors bring together many seemingly unrelated facts without providing a structure to help understand them. Chapter 4, the core of the book, emphasizes the use of microscopic principles in estimating rate parameters. The strength of this chapter is that it introduces collision and transition-state theories for both homogeneous and heterogeneous systems. This combined coverage, not usually done in kinetics texts, enables one to appreciate the analogy between reaction intermediates and surface complexes. Chapter 5, titled "Medium Effects," basically deals with the effect of energy exchange on reaction rates, but touches on surface inhomogeneity that is caused by lateral interactions between adsorbates. Chapter 6 attempts to integrate the concepts presented in the first five chapters and provides a microscopic viewpoint of heterogeneous catalysis. This chapter does provide some useful insights into surface-catalyzed reactions, but most of its sections are about kinetic modeling and not about application of the transition-state theory.

As an instructor I would consider using this book as a text in a graduate-level course that is devoted to kinetics. It needs to be supplemented with another book for a graduate-level course that is supposed to cover both kinetics and reaction engineering, as is commonly the case in many chemical engineering departments in the U.S. And despite its title, this book cannot be used alone in a special topic course on catalysis because there is no coverage on two key components of catalysis, namely synthesis and characterization. Readers who are interested in these two topics will need to turn to the other volumes in this series published by Plenum Press. Regardless of the nature of the course, this book's attractiveness as a text could be enhanced by the inclusion of homework problems and more extensive referencing. With some exceptions (mostly on tables and figures), all the references are now grouped together at the end of the book. This arrangement makes it difficult to do specific and selective follow-up reading, which an instructor may wish to do in preparation for class notes.

As a catalytic researcher I am happy to see the comprehensive and authoritative treatment of this book in providing a microscopic understanding of catalytic reactions. One topic that I would like to see more discussion on is kinetic modeling of surfaces that have a heterogeneous distribution of sites. Currently inhomogeneity is dealt with only in Section 5.7 within the context of adsorbate interactions. I would also welcome more examples showing the application of microscopic principles to industrially relevant reactions, or examples showing the development of new catalytic materials and technologies based on these principles. Such examples will demonstrate the power of microscopic theories as predictive tools as well as provide clear-cut procedures for applying them.

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La Combustion et les Flammes

By Roland Borghi and Michel Destriau, Editions Technip Paris cedex 15, 1995, 365 pp., 550 Fr. franc.

A new book, written in French by Roland Borghi of the University of Rouen and Michel Destriau of the University of Bordeaux, is a timely addition to the field of combustion literature. I cannot find a better way of characterizing the book than to translate the introductory remark of the publisher: "The book is intended primarily for students but also for engineers, professors, and researchers who desire to become acquainted with the field of combustion. Professionals in the field will find this book to be useful in view of the current global trend that takes into account mechanical, thermal and chemical aspects of engineering simultaneously."

After an introduction which gives a panorama of the applications of combustion in our modern society, the book starts with three chapters devoted to the fundamental aspects of combustion. Chapter 1 sets the base of the thermodynamics of combustion. Chapter 2 deals with chemical kinetics and introduces the notion of elementary reaction and of chain reaction. Chapter 3 is devoted to the physical phenomena of heat transfer and mass transfer. This chapter culminates with the demonstration and statement of the conservation equation of the aerothermochemistry. At this point, an original and instructive application of these equations to the famous diffusion-flame problem of Burke and Shumann is presented. The chapter ends with the generalization of the conservation equation to turbulent flow.

The road is now open to cover the theory of combustion and flames phenomena of various complexity. Chapter 4 deals with explosion in closed vessels. Chapter 5 considers flames in laminar flows, first premixed flames and then diffusion flames. These are mainly laboratory experiments for the study of flame structure and measurements of flame velocity. Chapter 6 on turbulent flames deserves special attention, since it is the field in which one of the authors (Borghi) is engaged actively in research. The chapter is specially well-documented and could also serve as a review article on the subject. Chapter 7 on the phenomena of detonation uses the classic approach of Chapman-Jouguet. Formation of tridimensional structure is discussed. The phenomenon of supersonic combustion is also treated in this chapter because of its similarities with detonation. Chapter 8 gives a fresh view on the problem of ignition based on recent work on computer simulation of the ignition process by various sources of energy.

Chapter 9 is concerned with the combustion of liquids and particle clouds. Having treated the classical problem of burning a single liquid drop, the authors discuss the structure and calculation of the flames of particle clouds. Advances in this new territory of research reveal exotic structures such as group combustion, percolant combustion, and combustion in pockets. The book ends with a chapter on environmental consideration regarding the chemistry of pollutants (NO_x, SO_x, and particulate).

This 365-page-long book is consistent with the stated purpose of the authors to write only an introductory text but with the maximum rigor and clarity possible. Although this book, according to the authors, emphasizes the physical aspect of the combustion phenomena rather than their mathematical demonstration, the mathematics gives the book its special flavor and makes it interesting enough to read or to study.

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Computational Modeling in Semiconductor Manufacturing

Edited by M. Meyyappan, Artech House, Norwood, MA, 1995, 363 pp., $89.00 (hardcover).

It has been recognized that computer simulation can be a valuable tool for the design and optimization of equipment used in the fabrication of microelectronic devices. This edited volume fills the need for a reference book on the subject. It is meant for researchers and graduate students with some background in semiconductor manufacturing processes. The book consists of six chapters: (1) modeling of semiconductor manufacturing processes—an introduction; (2) outline of numerical methods; (3) crystal growth; (4) chemical vapor deposition processes; (5) plasma process modeling; and (6) rapid thermal processing.

Chapter 1 (by M. Meyyappan) is a short introduction giving the rationale for and utility of numerical modeling as applied to semiconductor fabrication. Chapter 2 (by T. R. Govindan and F. J. de Jong) briefly discusses finite-difference techniques used to solve the partial-differential equation systems resulting by applying the principles of conservation of mass, momentum and energy. Emphasis is placed on the compressible Navier-Stokes equations for pure fluids. Discussed are the basics of finite difference approximations, applications to complex domains and nonuniform meshes, linearization of the equations, and iterative schemes to solve the resulting system of linear equations. The important issue of convergence acceleration is also touched on. Reference to other numerical techniques such as finite element and finite volume is made in passing, but these techniques are covered in Chapter 4 on chemical vapor deposition (CVD) processes.

Chapter 3 (by Y. T. Chan) that starts with a description of different techniques for crystal growth from vapor, melt, and solution focuses exclusively on growth from the melt. Governing equations and boundary conditions are presented, and the important dimensionless groups that arise are tabulated, and their physical meaning discussed. Criteria for stability of rotating flows, the onset of natural convection over vertical and horizontal surfaces, as well as convection due to surface tension gradients are presented. Stress analysis in crystal growth with explicit expressions for GaAs is also discussed. It is followed by case studies on some popular crystal growth techniques, which shed much insight into the factors that control the crystal diameter, shape of the melt/crystal interface, impurity incorporation into the crystal, and radial and axial uniformity of crystal properties. However, comparisons with experimental data are not made, except for one isolated case. The first case study deals with liquid encapsulated Czochralski (LEC) growth of GaAs crystals. Results of a 3-D simulation are shown, and the effect of varying the operating conditions is analyzed under quasi-steady state and transient thermal conditions. The effect of applying a static magnetic field B is reported, although the governing equations given earlier in the chapter do not include B. The float zone process is the second case study applied to growth of TiC. Here an account is given of the equations and procedure to determine the effect of a radio frequency (RF) heating system on the temperature distribution and the melt/solid interface shape. The last case study is a vertical Bridgman growth of HgCdTe. This chapter is well written, but the most important material system, silicon growth from the melt, is not discussed.

Chapter 4 (by C. R. Kleijn) is well written and thoroughly explains (thermal) CVD with an extensive literature review up to July 1993 (containing 290 references). Plasma CVD is dealt with in the next chapter. The author orients the reader with the basic principles of CVD, and common reactor designs and applications in the IC industry. An overview of the modeling approach is given followed by a detailed review of modeling assumptions, basic equations for mass, momentum and energy conservation in multicomponent mixtures, a general formalism for gas-phase and surface chemistry, and the relevant boundary conditions. Then empirical and kinetic theory results are given for calculating the thermodynamic and transport properties of multicomponent gas mixtures including thermodiffusion coefficients. Modeling of chemical reaction kinetics (including RRKM and transition state theories) and of conjugate heat transport are presented in a clear and concise manner. The author then presents an account of finite-volume and finite-element methods used to solve the governing equations. A comparison of the strengths and weaknesses of each technique, as well as a short discussion on the capabilities and shortcomings of available commercial CFD software packages, is also given. The author then tabulates a long list of CVD studies reported in the literature (up to July 1993) in a quite informative format including the publication year, process studied (e.g., CVD of Si and GaAs), problem dimensionality (2-D, 3-D, etc.), and type (diffusion, convective-diffusion, boundary layer, elliptic, parabolic, etc.), solution method (such as finite differences), and relevant features of the model (detailed vs. simplified chemistry, full multicomponent diffusion, detailed conjugate heat transfer, comparison with experiments, etc.).

He classifies the literature according to operating pressure and reactor configuration and presents a table for each classification. The author also gives an informative discussion of some of the most important papers in the corresponding classification, making comparisons with experimental data when possible. The classifications he considers are: atmospheric pressure CVD (APCVD) in horizontal rectangular ducts, APCVD in vertical impinging jet and stagnation point reactors, APCVD