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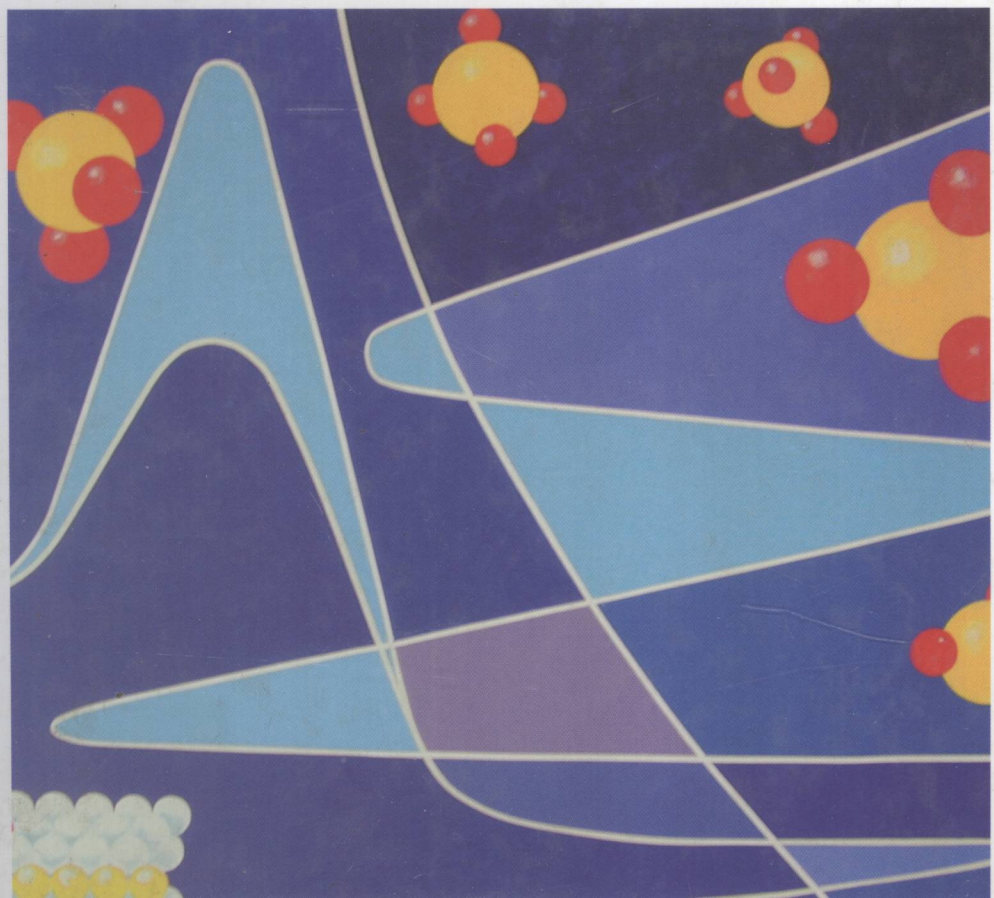
Concepts of Modern Catalysis and Kinetics

Second, Revised and Enlarged Edition



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Second, Revised and Enlarged Edition

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I. Chorkendorff, J. W. Niemantsverdriet
Concepts of Modern Catalysis and Kinetics

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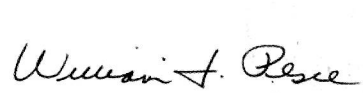

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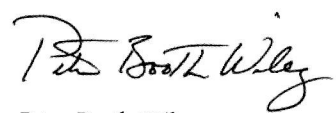

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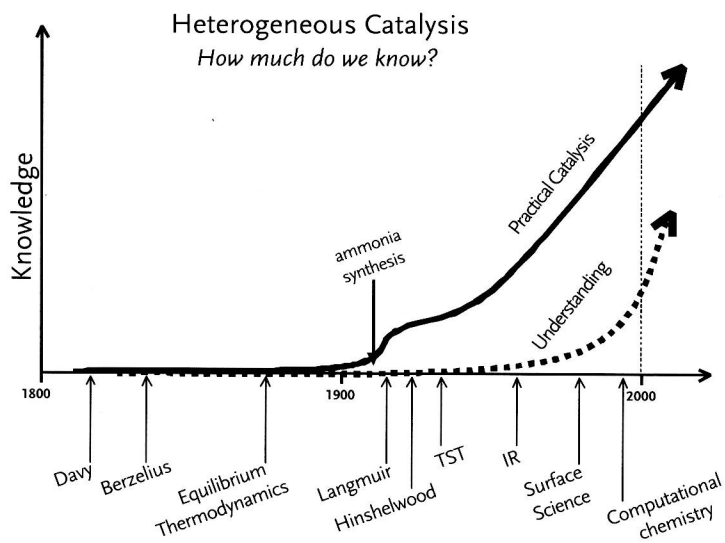
To Ina and Camilla

To Marianne, Hanneke,
Annemieke, Karin and Peter

Preface

Catalysis: Conceptually Understood But Far Away From Maturity

Catalysis as a phenomenon is becoming reasonably well understood on a conceptual level. Recognized as a phenomenon and utilized around 1816 by Davy in the mine safety lamp, and defined around 1835 by Berzelius, catalysis obtained an extensive empirical basis by the systematic experiments of Mittasch in the early 20th century. Studies of catalytic mechanisms became feasible when Langmuir – Hinshelwood kinetics became available in the mid 1920s. Since then, for many decades fundamental catalysis became more or less synonymous with kinetic analysis. The advent of spectroscopy, starting with infrared spectroscopy in the late 1950s, followed by a range of other techniques for catalyst characterization and investigation of surface species opened the opportunity to relate catalytic properties with composition and structure of materials. Surface science enabled one to resolve adsorption geometries and reactivity patterns in well-defined structures, culminating in scanning tunneling spectroscopy as the ultimate tool to resolve surface structure and adsorbed spe-



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cies with atomic precision by the end of the 20th century. Techniques have become available to study catalysts under working conditions. Ever increasing computational power enables the calculation of adsorbate geometries, bond strengths and even reaction rates. Anno 2003, catalysis has developed into a scientific discipline with a firm conceptual basis. The relation between catalytic activity for a certain reaction and the composition, structure of a surface is in general qualitatively well understood, provided the surfaces are relatively simple and defined.

However, the prospect of designing a catalyst from first principles behind our desk is still far away. A number of reasons is responsible. Although we may be able to describe a catalytic reaction on a well-defined single crystal of a metal under well-defined and simplified conditions, this becomes tremendously more complicated when the same reaction runs over small catalyst particles on a support in a realistic reactor environment. The world of the ideal surface science laboratory and that of industrial practice are not only separated by the often cited pressure gap, but also by structural, and materials gaps. The complexity of small, supported crystallites that dynamically respond to every change in reaction environment is only beginning to be explored. Secondly, the way we describe the kinetics of catalytic reactions is, albeit greatly refined, still based on the adsorption isotherm of Langmuir (1915) and the kinetic formalism of Hinshelwood (1927), based on ideal surfaces with equivalent adsorption sites and adsorbate species that are randomly mixed and do not interact. This represents another gross over simplification, which has recently become recognized and is beginning to become addressed in terms of ensembles with reactivities many orders of magnitude more reactive than the planar surfaces.

This book intends to be an introduction to the fundamentals of heterogeneous catalysis, aiming to explain the phenomenon of catalysis on a conceptual level. Kinetics, being the tool to investigate and describe catalytic reactivity as it expresses itself in a reactor, plays an important role in this book. Also reaction rate theory, providing the formalism to relate reaction rates to molecular structure of reacting species is described extensively. Next we describe catalytic surfaces, as well as the tools to study them. With this knowledge we treat surface reactivity in greatly simplified molecular orbital theory, again with the aim to give a conceptual explanation of how a catalyst works and what are the trends when going from one surface/structure to another. The final chapters serve to illustrate catalysis in practice, to give the reader an impression of how catalysis is applied. We emphasize that the book is a textbook, written for students in chemistry, physics and chemical engineering who are interested in understanding the concepts. Many more important details are worthwhile knowing, but are beyond the scope of this book and will have to be found in specialist literature.

This book is based on courses, which the authors have taught at Lyngby and Eindhoven for many years. For example, Chapters 1 to 3 form the basis for a mandatory course "Kinetics and Catalysis" presented in the second year of the Bachelor's curriculum at Eindhoven, while Chapters 4, 5 and 8 to 10 formed the basis for an optional course Introduction to Catalysis. In Lyngby, Chapters 1 to 7 have been used for an optional course in "Chemical Reaction Kinetics and Catalysis" in the Master's curriculum. At the end of the book we have added a list of questions for every chap-

ter, which students may use to test their knowledge. The exercises are mainly meant to enable students to acquire skills in kinetic modelling. Some of these exercises have been used in written examinations. We intend to make solutions available on our web sites (www.cinf.dtu.dk and www.catalysis.nl).

The authors are indebted to many colleagues in the field of catalysis and surface science. We mention in particular Jens Nørskov and Rutger van Santen. Both made very substantial contributions to the theory of heterogeneous catalysis. We are very grateful for the many extensive and pleasant discussions we had with them. Also our contacts with industry have been most inspiring and indispensable. We mention in particular Haldor Topsøe A/S in Lyngby, the Shell Research and Technology Centre in Amsterdam and SASOL Research in South Africa. We also want to thank the numerous students who followed our courses in Lyngby and Eindhoven. They have taught us more than they perhaps realize. Finally we thank our families who patiently allowed us to spend a considerable amount of time on this book. We devote this book to them.

Ib Chorkendorff
Hans Niemantsverdriet

List of Acronyms

AES	Auger Electron Spectroscopy
AFM	Atomic Force Microscopy
bcc	Body-centered-cubic
BET	Brunauer–Emmet–Teller
CSTR	Continuously Stirred Tank Reactor
DFT	Density Functional Theory
DFT-GGA	Density Functional Theory Generalized Gradient Approximation
DOS	Density Of States
EDX	Energy Dispersive X-ray Analysis
ER	Eley-Rideal
ESR	Electron Spin Resonance
EXAFS	Extended X-ray Absorption Fine Structure
FCC	Fluidized Catalytic Cracking
fcc	face centered cubic
hcp	hexagonally close packed
HDA	Hydrodearomatization
HDM	Hydrodemetallization
HDO	Hydrodeoxygenation
HDN	Hydrogdenitrogenation
HDS	Hydrodesulfurization
HOMO	Highest Occupied Molecular Orbital
IR	Infra Red
IRAS	Infra Red Absorption Spectroscopy
ISS	Ion Scattering Spectroscopy
LCAO	Linear Combination of Atomic Orbitals
LEED	Low Energy Electron Diffraction
LEIS	Low Energy Ion Scattering
LH	Langmuir Hinshelwood
LPG	Liquefied Petroleum Gas
LUMO	Lowest Unoccupied Molecular Orbital
MARI	Most Abundant Reaction Intermediate
MTBE	Methyl Tertiary Butyl Ether
NMR	Nuclear Magnetic Resonance

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P	Product
PEMFC	Proton Exchange Membrane Fuel Cell
PFR	Plug Flow Reactor
R	Reactant
RBS	Rutherford Back Scattering
RLS	Rate Limiting Step
RON	Research Octane Number
RWGS	Reverse Water gas Shift
SATP	Standard Ambient Temperature and Pressure
SCR	Selective Catalytic Reduction
SEM	Scanning Electron Microscopy
SIMS	Secondary Ion Mass Spectroscopy
SMSI	Strong Metal Support Interaction
SOFC	Solid Oxide Fuel Cell
SPA-LEED	Spot Profile Analysis Low Energy Electron Diffraction
SPARG	Sulfur Passivated Reforming Process
STM	Scanning Tunneling Microscopy
STP	Standard Temperature and Pressure
TEM	Transmission Electron Microscopy
TPD	Temperature programmed Desorption
TPH	Temperature programmed Hydrogenation
TPO	Temperature programmed Oxidation
TPR	Temperature programmed Reaction
TPS	Temperature programmed Sulfidation
TS	Transition State
TWC	Three Way Catalyst
UHV	Ultra High Vacuum
UPS	Ultra Violet Electron Spectroscopy
UV-VIS	Ultra Violet and Visible Absorption
WGS	Water Gas Shift
XANES	X-ray Absorption Near Edge Spectroscopy
XPS	X-ray Photoelectron Spectroscopy
XRD	X-ray Diffraction
YSZ	Yttrium Stabilized Zirconium

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