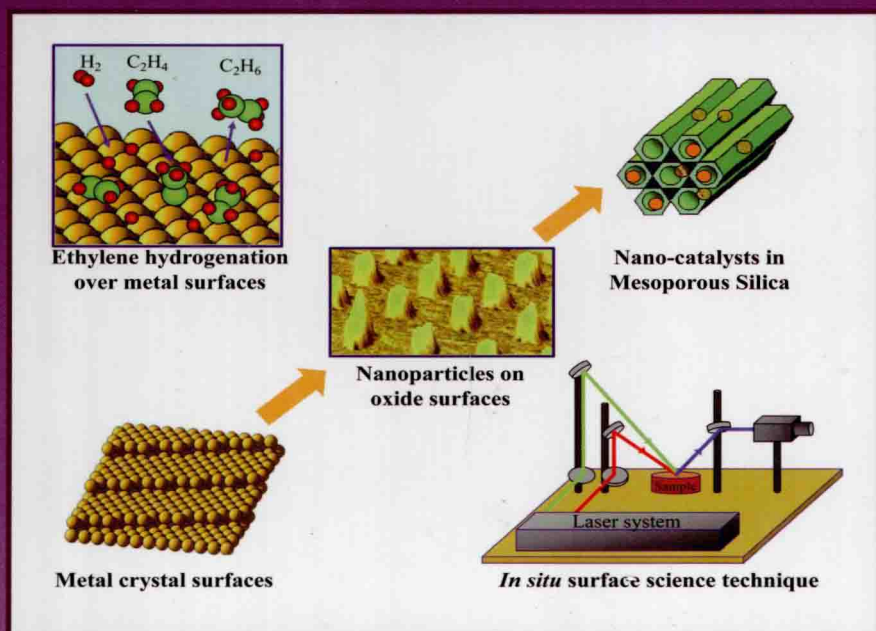


# INTRODUCTION TO SURFACE CHEMISTRY AND CATALYSIS

SECOND EDITION



Gabor A. Somorjai  
Yimin Li

# INTRODUCTION TO SURFACE CHEMISTRY AND CATALYSIS

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Second Edition

GABOR A. SOMORJAI

YIMIN LI

Department of Chemistry, University of California, Berkeley, California



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**INTRODUCTION TO  
SURFACE CHEMISTRY  
AND CATALYSIS**

*To my grandchildren Stephanie, Clara, Benjamin, and Diana*

Gabor A. Somorjai

*To my wife Hailan and my children Xinyue and Xincheng*

Yimin Li

# PREFACE

The chemistry of surfaces and interfaces plays dominating roles in our lives. The challenge of modern physical chemistry is to understand macroscopic surface phenomena on the molecular level. This book, the second edition of *Introduction to Surface Chemistry and Catalysis*, first published in 1994, aims to update our rapid progress in these fields. In the interim period, surface science has emerged as the frontier area of molecular physical chemistry on a broad front, ranging from nanoparticle structures to biointerfaces and selective catalysis of stereospecific molecules and reactions, and to chemical energy conversion. Many applications of the newly acquired knowledge of molecular surface chemistry is used in innovative technologies relying on metal, semiconductor, and polymer surfaces in order to achieve controlled chemical bonding, adhesion, friction, electron and atom transport, solar energy conversion, and selective catalysis.

Since the early 1960s, techniques for the study of surfaces on the molecular level, which provided the foundation for the rapid evolution of surface science, have become available in ever-increasing numbers. Model surfaces, mostly metals and semiconductors, were utilized first in the form of single crystals. This enabled us to control the atomic surface structure, which is an important factor in influencing surface chemistry and the formation of surface chemical bonds. The aim was to maintain atomic level control of structural complexity wherever possible. The dominant application of surface chemistry is in catalysis. Indeed, most catalytic systems (heterogeneous, homogeneous, and enzymatic) are nanoparticles and so metal nanoparticles well represent working catalytic systems. Recently, nanoparticles in the 1–10-nm range of uniform size and shape have been prepared and then used as model catalyst systems. Previously, in the 1960s and 1970s, methods using ultrahigh vacuum provided the only sure way to prepare atomically clean surfaces and avoid contamination that would modify their chemistry. Most studies were carried out over clean surfaces using mostly low-energy electron diffraction–surface crystallography, electron spectroscopies, and molecular probes of adsorption and desorption at low pressures. Because most applications of surface chemistry occur at high ambient pressures at solid–gas, solid–liquid, and solid–solid interfaces, techniques were developed at Berkeley that could monitor

surface properties at the molecular level at the “buried” interfaces. These were sum frequency generation—surface vibrational spectroscopy, high-pressure scanning tunneling microscopy, and ambient pressure X-ray photoelectron spectroscopy. With the help of many other techniques developed in other laboratories, the molecular science of surfaces and its many applications to chemical, biochemical, mechanical, electrical, optical, and magnetic properties could be investigated. Consequently, many new phenomena were discovered, and the concepts that make interfaces a special media were identified. The new molecular understanding spawned many new applications. Our focus of investigation was the structural and chemical surface properties and their atomic and molecular scale behavior when applied to chemical bonding, catalysis, and biointerfaces. This second edition reviews the rapid developments of molecular physical chemistry of surfaces in these fields during the past 15 years and complements the knowledge that was accumulated and described in the first edition.

We have had the good fortune to work with outstanding students and postdoctoral fellows. Their research accomplishments appear on virtually every page of this book. The Department of Chemistry, the Berkeley campus of the University of California, and the Lawrence Berkeley National Laboratory have provided an ideal environment to carry out research because of their intense but friendly atmosphere, and constant pursuit of excellence. We learned much of what we know from our colleagues and students in Berkeley. The Department of Energy, through its Basic Energy Sciences, provides most of the funding (for which we are most grateful) for our research through the Lawrence Berkeley National Laboratory.

We would like to acknowledge our colleagues throughout the world who, through their research in the field of surface science and catalysis, contributed to this book. Their results are displayed in the text and in many of the figures and tables. We would like to thank Professor Herbert Strauss and graduate students Derek Butcher, J. Russell Renzas, and George Holinga for their careful reading and editing of the manuscript. We appreciate the advice and help of Inger Coble. We are grateful to Professor Fabio H. Ribeiro for providing us with updated kinetic data of catalyzed hydrocarbon reactions, and we would like to thank Stephanie Burt, Marijke van Spyk, Hyun Hwa Jo, and Dr. Ruxia Jiang for their assistance in the preparation of references and tables. Last but not least, we would like to thank the editorial staff of John Wiley & Sons, Inc., for the final editing.

GABOR A. SOMORJAI  
YIMIN LI

*Berkeley, California*  
*February 8, 2010*

### Fundamental Constants

Constant	Symbol	Value
Speed of light	$c$	$2.998 \times 10^{10}$ cm/sec = $2.998 \times 10^8$ m/sec
Planck's constant	$h$	$6.626 \times 10^{-27}$ erg · sec = $6.626 \times 10^{-34}$ J · sec
Avogadro's number	$N_A$	$6.022 \times 10^{23}$ molecules/mole
Electron charge	$e$	$1.602 \times 10^{-21}$ coulombs = $4.803 \times 10^{-10}$ esu
Gas constant	$R$	$1.987$ cal/deg/mole = $8.315$ J/deg/mole
Boltzmann's constant	$k_B$	$1.381 \times 10^{-16}$ erg/deg = $1.381 \times 10^{-23}$ J/deg = $R/N_A$
Gravitational constant	$g$	$9.807$ m/sec <sup>2</sup>
Permittivity of vacuum	$\epsilon_0$	$8.854 \times 10^{-12}$ C <sup>2</sup> /J/m

### Prefixes

Z	zepto	$10^{-21}$
a	Atto	$10^{-18}$
f	femto	$10^{-15}$
p	pico	$10^{-12}$
n	nano	$10^{-9}$
$\mu$	micro	$10^{-6}$
m	milli	$10^{-3}$
c	centi	$10^{-2}$
d	deci	$10^{-1}$
da	deca	$10^1$
k	kilo	$10^3$
M	mega	$10^6$
G	giga	$10^9$
T	tera	$10^{12}$
P	peta	$10^{15}$

### Several Length Units

Name of unit	Symbol	Definition	Relation to SI units
meter (SI base unit)	m	$\equiv$ Distance light travels in 1/299 792 458 of a second in vacuum	$= 1$ m
ångström	Å	$\equiv 1 \times 10^{-10}$ m	$= 10^{-10}$ m = 0.1 nm
bohr, atomic unit of length	$a_0$	$\equiv$ Bohr radius of hydrogen	$= 5.292 \times 10^{-11}$ m = 0.5292 Å



Force Conversion Table

	newton (N)	dyne (dyn)	kilogram-force (kp)
N	$\equiv 1 \text{ kg} \cdot \text{m/s}^2$	$10^5$	0.10197
dyn	$10^{-5}$	$\equiv 1 \text{ g} \cdot \text{cm/s}^2$	$1.0197 \times 10^{-6}$
kp	9.80665	980665	$\equiv \text{g} \cdot (1 \text{ kg})$

Example reading:  $1 \text{ N} = 10^5 \text{ dyn} = 0.1097 \text{ kp}$ .

Pressure Conversion Table

	pascal (Pa)	bar (bar)	technical atmosphere (at)	atmosphere (atm)	torr (Torr)
1 Pa	$\equiv 1 \text{ N/m}^2$	$10^{-5}$	$1.0197 \times 10^{-5}$	$9.8692 \times 10^{-6}$	$7.5006 \times 10^{-3}$
1 bar	100,000	$\equiv 10^6 \text{ dyn/cm}^2$	1.0197	0.98692	750.06
1 at	98,066.5	0.980665	$\equiv 1 \text{ kgf/cm}^2$	0.96784	735.56
1 atm	101,325	1.01325	1.0332	$\equiv 1 \text{ atm}$	760
1 torr	133.322	$1.3332 \times 10^{-3}$	$1.3595 \times 10^{-3}$	$1.3158 \times 10^{-3}$	$\equiv 1 \text{ Torr};$ $\approx 1 \text{ mmHg}$

Example reading:  $1 \text{ Pa} = 1 \text{ N/m}^2 = 10^{-5} \text{ bar} = 10.197 \times 10^{-6} \text{ at} = 9.8692 \times 10^{-6} \text{ atm}$ , etc.

Energy Conversion Table

	erg	eV	$\text{cm}^{-1}$	kcal/mol	kJ/mol	K	J	Hz
erg	1	$6.241 \times 10^{11}$	$5.034 \times 10^{15}$	$1.44 \times 10^{13}$	$6.02 \times 10^{13}$	$7.243 \times 10^{15}$	$10^{-7}$	$1.509 \times 10^{26}$
eV	$1.602 \times 10^{-12}$	1	$8.066 \times 10^3$	23.06	96.49	$1.160 \times 10^4$	$1.602 \times 10^{-19}$	$2.418 \times 10^{14}$
$\text{cm}^{-1}$	$1.986 \times 10^{-16}$	$1.240 \times 10^{-4}$	1	$2.859 \times 10^{-3}$	$1.196 \times 10^{-2}$	1.439	$1.986 \times 10^{-23}$	$2.998 \times 10^{10}$
kcal/mol	$6.948 \times 10^{-14}$	$4.336 \times 10^{-2}$	$3.498 \times 10^2$	1	4.184	$5.032 \times 10^2$	$6.948 \times 10^{-21}$	$1.049 \times 10^{13}$
kJ/mol	$1.661 \times 10^{-14}$	$1.036 \times 10^{-2}$	83.59	0.2390	1	$1.203 \times 10^2$	$1.661 \times 10^{-21}$	$2.506 \times 10^{12}$
K	$1.381 \times 10^{-16}$	$8.617 \times 10^{-5}$	0.6950	$1.987 \times 10^{-3}$	$8.314 \times 10^{-3}$	1	$1.381 \times 10^{-23}$	$2.084 \times 10^{10}$
J	$10^7$	$6.242 \times 10^{18}$	$5.034 \times 10^{22}$	$1.44 \times 10^{20}$	$6.02 \times 10^{20}$	$7.243 \times 10^{22}$	1	$1.509 \times 10^{33}$
Hz	$6.626 \times 10^{-27}$	$4.136 \times 10^{-15}$	$3.336 \times 10^{-11}$	$9.537 \times 10^{-14}$	$3.990 \times 10^{-13}$	$4.799 \times 10^{-11}$	$6.626 \times 10^{-34}$	1

Example reading:  $1 \text{ eV} = 1.602 \times 10^{-12} \text{ erg} = 8.066 \times 10^3 \text{ cm}^{-1} = 23.06 \text{ kcal/mole} = 96.49 \text{ kJ/mole}$ , etc.

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# INTRODUCTION

The purpose of this book is to describe the present state of development of modern surface science at an introductory level to students of the physical sciences and engineering. Junior standing in chemistry, physics, engineering, or the life sciences would qualify the student to take a course that would make use of this text. Teachers of the introductory general chemistry courses which are usually given during the first year of university or college enrollment, could use certain chapters (with deletions of some of the derivations) to supplement discussions of thermodynamics or catalysis, for example. We have used some of the chapters as supplementary material in our freshman and our core physical chemistry courses at Berkeley. The book should also be useful as a reference for professionals in need of data and concepts related to the properties of surfaces and interfaces.

Since the publication of the first edition in 1994, molecular surface chemistry has developed rapidly and has expanded into fields where atomic scale and molecular information were previously not available. This second edition reflects this impressive increase of information in virtually every chapter. A new chapter has been added on “soft” surfaces, polymers, and biointerfaces. Chapter 1, Surfaces: an Introduction, offers a brief history of the evolution of surface chemistry and a classification of properties of surfaces and interfaces, including clusters and small particles, where the science of nanoparticles has shown impressive gains. The compilation of techniques available for atomic and molecular level studies of surfaces has been extended to include the newly developed instruments that are now available for surface studies. One of the major uses of a textbook is to provide experimental data of properties, and we have updated the database in each chapter to include the rapid increase of information over the last 15 years. Chapter 2 discusses the structure of clean and adsorbate-covered surfaces and the ordered defects and adsorbates, reconstruction, and

adsorbate-induced restructuring that are structural properties unique to surfaces. Chapter 3 focuses on the equilibrium thermodynamic properties of surfaces, including the properties of curved surfaces. Thermodynamics of adsorbed monolayers are also included. The motion of surface atoms about their equilibrium positions and along the surface (i.e., diffusion) is reviewed in Chapter 4. Energy transfer during gas–surface interactions and discussions of elementary reaction steps, adsorption, surface diffusion, and desorption are part of this chapter. The electrical properties of surfaces is the subject of Chapter 5. The electron density distribution at the surface that gives rise to the surface electron potential and the interface dipole layer is introduced and the effects of interface properties and adsorption on the contact potentials and work function are discussed. The emission of valence and inner-shell electrons, surface ionization, and various surface science techniques based on electron diffraction, excitation, and tunneling are introduced in this chapter. Chapter 6 focuses on the nature of the surface chemical bond. Discussed are the bonding trends on transition metal surfaces across the Periodic Table and the theory that explains how the filling of the d-band with electrons dominates the adsorbate bonding. The carbon monoxide metal surface chemisorption bond provides a detailed picture of adsorbate molecular bonding at surfaces. Many important surface bonding concepts, including adsorbate-induced surface restructuring, thermal activation of bond breaking, surface structure sensitivity, and coverage dependence of bonding are reviewed. Discussions of weak surface bond and physical adsorption are included. Chapter 7 is devoted to mechanical properties of surfaces. Basic concepts of elastic modulus, yield strength, dislocations, deformation, hardness, adhesion, and friction force are introduced. However, our focus is on the understanding of surface mechanical properties at the molecular level. The information obtained from atomic force microscopy (AFM) studies are reviewed along with the new field of nanotribology and single bond stretching studies. The case history of diamond is reviewed. Polymer surfaces and biointerfaces are introduced in Chapter 8. This field has developed rapidly in the last 15 years. Polymer surface structure and glass transition are discussed. Surface segregation of bulk additives, side groups, and end-groups as they control polymer surface properties is introduced. The great importance of biointerfaces in biotechnology is demonstrated. The adsorption of proteins, peptides, and amino acids on surfaces and their bonding interactions are reviewed. The final chapter (Chapter 9) is devoted to catalysis by surfaces. A brief history and concepts of catalysis are reviewed followed by discussions of catalyst preparation, techniques of characterization, and methods of reaction studies. *In situ* characterization techniques are emphasized and concepts of metal catalysis are discussed. We then focus on case histories of surface catalysis, ranging from ethylene hydrogenation and ammonia synthesis to the oxidation of carbon monoxide. Detailed discussions of these one-product reactions are followed by a review of selectivity in multi-path reactions; how the surface structure, nanoparticle size, bimetallic systems, metal-support interactions, oxidation states, reaction intermediates, and reaction conditions influence reaction selectivity.

All through this book, the concepts of surface chemistry as they emerge from experimental studies of surfaces are emphasized. A summary at the end of each chapter reviews the important conclusions of studies that were discussed. As we focus on molecular surface properties that emerged from surface science studies, there are important areas of surface chemistry and catalysis that have not been covered. These omissions were made for two reasons: (1) we primarily discuss local atomic and molecular properties of surfaces, structure, chemical bonding, mechanical properties, and chemical reactivity; transport properties of surfaces (e.g., optical, electron transport, and magnetic properties) are not

discussed reflecting the interests of the authors and the availability of excellent reviews and books that cover these subjects. (2) When we avoid discussions of case histories of other systems that are in the literature, we plead for forgiveness. We believe the reader would not have benefited from broader exposure to more systems beyond these few needed to introduce surface phenomena and concepts for the sake of clarity and focus.



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# 1

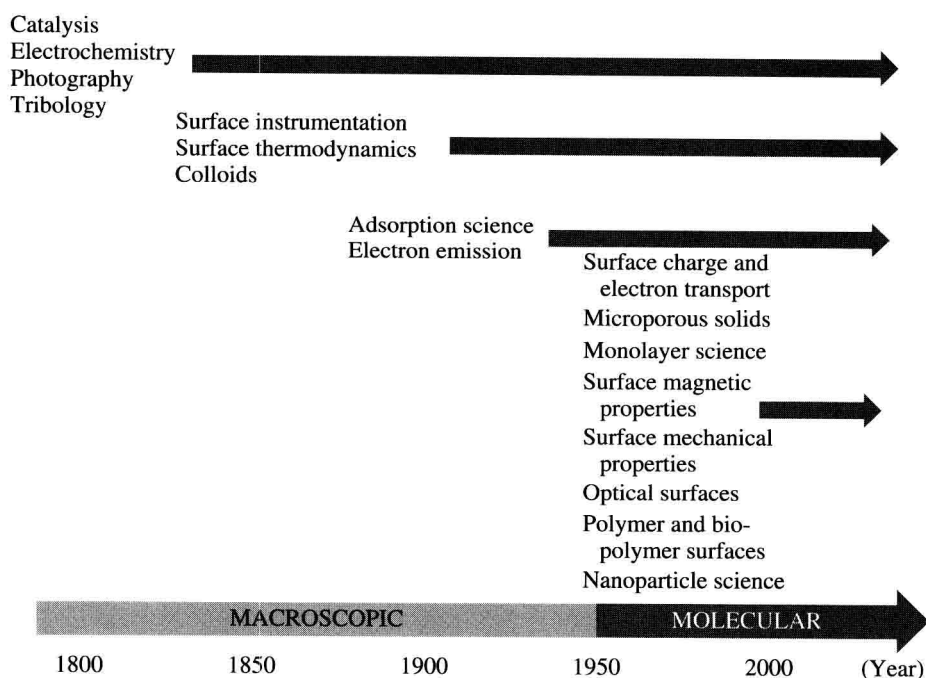
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### 1.1 HISTORICAL PERSPECTIVE

Surface science in general and surface chemistry in particular have a long and distinguished history. The spontaneous spreading of oil on water was described in ancient times and was studied by Benjamin Franklin. A timeline of the historical development of surface chemistry since then is shown in Figure 1.1. The application of catalysis started in the early 1800s, with the discovery of the platinum (Pt)-surface-catalyzed reaction of  $\text{H}_2$  and  $\text{O}_2$  in 1823 by Dobereiner. He used this reaction in his “lighter” (i.e., a portable flame) source, of which he sold a large number. By 1835 [1], the discovery of heterogeneous catalysis was complete thanks to the studies of Kirchhoff, Davy, Henry, Philips, Faraday, and Berzelius. It was at about this time that the Daguerre process was introduced for photography. The study of tribology, or friction, also started around this time, coinciding with the industrial revolution,



**Figure 1.1.** Timeline of the historical development of surface chemistry.

although some level of understanding of friction appears in the work of Leonardo da Vinci. Surface-catalyzed chemistry-based technologies first appeared in the period of 1860–1912, starting with the Deacon process ( $2\text{HCl} + \text{O}_2 \rightarrow \text{H}_2\text{O} + \text{Cl}_2$ ),  $\text{SO}_2$  oxidation to  $\text{SO}_3$  (Messel, 1875), the reaction of methane ( $\text{CH}_4$ ) with steam to produce  $\text{CO}$  and  $\text{H}_2$  (Mond, 1888), ammonia ( $\text{NH}_3$ ) oxidation (Ostwald, 1901), ethylene ( $\text{C}_2\text{H}_4$ ) hydrogenation (Sabatier, 1902), and  $\text{NH}_3$  synthesis (Haber, Mittasch, 1905–1912). Surface tension measurements and recognition of equilibrium constraints on surface-chemical processes led to the development of the thermodynamics of surface phases by Gibbs (1877). The existence of polyatomic or polymolecular aggregates that lack crystallinity and diffuse slowly (e.g., gelatine and albumin) was described in 1861 by Graham, who called these systems “colloids”. Polymolecular aggregates that exhibit internal structure were called “micelles” by Nageli, and stable metal colloids were prepared by Faraday. However, the colloid subfield of surface chemistry gained prominence at the beginning of the 20th century with the rise of the paint industry and the preparation of artificial rubbers. Studies of light bulb filament lifetimes, high-surface-area gas absorbers in the gas mask, and gas-separation technologies in other forms, led to investigations of atomic and molecular adsorption (Langmuir, 1915). The properties of chemisorbed and physisorbed monolayers, adsorption isotherms, dissociative adsorption, energy exchange, and sticking upon gas–surface collisions were studied. Studies of electrode surfaces in electrochemistry led to the detection of the surface space charge [2] (for a review of electrochemistry in the 19th century, see Ref. [3]). The surface diffraction of electrons was discovered by Davisson and Germer (1927). Major academic and industrial laboratories focusing on surface studies have been formed in Germany (Haber, Polanyi, Farkas, Bonhoefer), the United Kingdom (Rideal, Roberts, Bowden), the



United States (Langmuir, Emmett, Harkins, Taylor, Ipatief, Adams), and many other countries. They have helped to bring surface chemistry into the center of development of chemistry—both because of the intellectual challenge to understand the rich diversity of surface phenomena and because of its importance in chemical and energy conversion technologies.

In the early 1950s, focus in chemistry research shifted to studies of gas-phase molecular processes, as many new techniques were developed to study gas-phase species on the molecular level. This was not the case in surface and interface chemistry, although the newly developed field-ion and electron microscopies did provide atomic level information on surface structure. The development of surface-chemistry-based technologies continued at a very high rate, however, especially in areas of petroleum refining and the production of commodity chemicals. Then, in the late 1950s, the rise of the solid-state-device-based electronics industry and the availability of economical ultrahigh vacuum systems (UHV) (developed by research in space sciences) provided surface chemistry with new challenges and opportunities, resulting in an explosive growth of the discipline. Clean surfaces of single crystals could be studied for the first time, and the preparation of surfaces and interfaces with known atomic structure and controlled composition was driving the development of microelectronics and computer technologies. New surface instrumentation and techniques have been developed that permit the study of surface properties on the atomic scale. Many of the most frequently used surface characterization techniques are listed in Table 1.1. Most of these have been developed since the 1960s.

As a result of the sudden availability of surface characterization techniques, macroscopic surface phenomena (adsorption, bonding, catalysis, oxidation and other surface reactions, diffusion, desorption, melting and other phase transformation, growth, nucleation, charge transport, atom, ion, and electron scattering, friction, hardness, lubrication) are being re-examined on the molecular scale. This finding has led to a remarkable growth of surface chemistry that has continued uninterrupted to date. The discipline has again become one of the frontier areas of chemistry. The newly gained knowledge of the molecular ingredients of surface phenomena has given birth to a steady stream of high-technology products, including new hard coatings that passivate surfaces; chemically treated glass, semiconductor, metal, and polymer surfaces, where the treatment imparts unique surface properties; newly designed catalysts, chemical sensors, and carbon fiber composites; surface-space-charge-based copying; and new methods of electric, magnetic, and optical signal processing and storage. Molecular surface chemistry is being utilized increasingly in biological science.

## 1.2 SURFACES AND INTERFACES: CLASSIFICATION OF PROPERTIES

Condensed phases (solids and liquids) must have surfaces or interfaces. The suit of an astronaut maneuvering in outer space represents a solid–vacuum interface (Fig. 1.2a); a basketball player jumping to score is a moving solid–gas interface (Fig. 1.2b); a sailboat moving over the waves is a solid–liquid interface (Fig. 1.2c); a tire sliding at the solid–solid interface (Fig. 1.2d). The surface of a lake is a liquid–gas interface. Olive oil poured on top of an open bottle of wine to prevent air oxidation forms a liquid–liquid interface. These interfaces exhibit some remarkable physical and chemical properties. The chemical behavior of surfaces is responsible for heterogeneous catalysis (e.g.,  $\text{NH}_3$  synthesis) and gas separations (as in the extraction of oxygen and nitrogen from air) by selective adsorption. Mechanical surface