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Preface to the Molecular Surface Chemistry and Its Applications Special Issue

This issue of *Langmuir* is a celebration of the scientist, leader, and friend Professor Gabor A. Somorjai on the occasion of his 75th birthday. This issue is very fitting, since Professor Somorjai is recognized for laying the foundation of modern surface chemistry and has made advances of the same magnitude as those of Irving Langmuir himself. One of his main contributions is in the area of heterogeneous catalytic chemistry which he converted from an empirical art into a science whereby catalytic processes can be quantified and the catalytic site can be understood in terms of surface atomic structures and fundamental molecular properties. Over the past 45 years, he shared his knowledge and enthusiasm with students and collaborators who now work on the many ideas and concepts he created. His hallmark is to always evolve and never stay with the same technique or reaction for too long. He is always breaking new ground. He began with studies of catalytic surfaces under ultrahigh vacuum but evolved to study reactions at solid–liquid interfaces and to studying surface processes at practical conditions. This issue is a reflection of the breadth and depth of his contributions to surface science.

Biographical Summary

Gabor A. Somorjai was born in Budapest, Hungary, in 1935. He started his journey in chemistry at the Technical University of Budapest as a chemical engineering student in 1953. In the fall of 1956, the Hungarian Revolution broke out with the active participation of the students in the Technical University (including himself), but it soon failed when the Soviet military occupied Budapest. He decided to leave Hungary and walked across the border to Austria with his sister and girlfriend, Ms. Judith Kaldor who later became his wife. He was a few months from getting his diploma when in 1957, he immigrated to the United States.

While at Camp Kilmer in New York, he wrote a number of letters to universities in the United States, inquiring about graduate studies. He was admitted to the University of California at Berkeley as a graduate student. At the time, he was interested in studying either heterogeneous catalysis or polymers. The closest project he could find at that time was offered by Professor Richard Powell, a professor of inorganic chemistry at UC Berkeley. He conducted small-angle X-ray scattering to characterize the particle size and size distribution of platinum nanoparticles on alumina catalysts provided by Chevron (Standard Oil of California at that time). He earned his Ph.D. degree in 1960 and then joined the research staff at IBM Research in New York. In 1962, while he was working for IBM, he became a United States citizen.

At IBM, he investigated the properties of cadmium sulfide and other luminescent materials. Around that time, he witnessed the development of low energy electron diffraction (LEED) at Bell Laboratories and recognized the power and importance of this technique. He purchased the first LEED unit commercially sold from Varian and began his studies in basic surface science. Later, IBM promoted him to a manager position, but he did not like losing the opportunity of conducting fundamental science of interest to him. He decided to leave IBM and join the faculty of the Chemistry Department at Berkeley as an assistant professor in 1964. When he joined the faculty, the Inorganic Materials Research Division (IMRD) of the Lawrence Berkeley National Laboratories (LBNL) was established, and he was offered a position as a Principal Investigator in IMRD.

At Berkeley, he launched a research program focusing on surface chemistry and physics using LEED. Not surprisingly, his first choice of material was platinum—a typical catalyst used in the petroleum refining industry and the material he studied during his Ph.D. at Berkeley. He realized the importance of detailed surface structure to understand the mechanism of heterogeneous catalysis and devoted his efforts in unveiling the surface structure of clean crystal surfaces and those covered with chemisorbed organic species. This was the genesis of modern surface chemistry on model catalysts. He was promoted to Associate Professor in 1967 and to Professor in 1972. In 2002, the UC Board of Regents appointed him as University Professor of the UC system. He is also a Faculty Senior Scientist in the Materials Sciences Division and Director of the Surface Science and Catalysis Program at the Center for Advanced Materials, at LBNL.

Professor Somorjai is the author of nearly 1100 scientific papers in the fields of surface chemistry, heterogeneous catalysis, solid state chemistry, polymers, and biomaterials. He has written three textbooks and one monograph.

Honors and Awards

He received numerous honors and awards: Guggenheim Fellow (1969); Unilever Professor, University of Bristol (1972); Kokes Award, Johns Hopkins University (1976); Emmett Award of the American Catalysis Society (1977); Miller Professor, Berkeley (1978); Member, National Academy of Sciences (1979); Colloid and Surface Chemistry Award of the American Chemical Society (1981); Member, American Academy of Arts and Sciences (1983); Henry Albert Palladium Medal (1986); Peter Debye Award in Physical Chemistry of the American Chemical Society (1989); Adamson Award in Surface Chemistry of the American Chemical Society (1994); Van Hippel Award of the Materials Research Society (1997); Wolf Prize in chemistry (1998); Creative Research Award in Homogeneous and Heterogeneous Catalysis of the American Chemical Society (2000); Linus Pauling Award (2000); National Medal of Science (2002); Remsen Award from the Maryland Section of the American Chemical Society, (2006); Langmuir Prize from the American Physical Society (2007); Priestley Medal (2008), Japanese Society for the Promotion of Science Award (2009); Excellence in Surface Science Award from the Surfaces in Biointerfaces Foundation (2009); Fellowship of the American Chemical Society (2009); and Honorary Membership, Chemical Society of Japan (2009).

Summary of Research Contributions

Somorjai started his work in 1965 by determining, under ultrahigh vacuum, the surface structure of clean single platinum crystals cut along definite crystal axes.[1] His first major discovery was that the atomic structure of metal surfaces is different from the structure of the body of the solid, and he found that the surface structure changes when other molecules are adsorbed on it.[1,2] One of Somorjai's early discoveries has entered physical chemistry textbooks, and it is an essential mental tool for those who design or improve practical catalysts.

He found that the surface of a pure single crystal, even one cut along the (100) plane, is not absolutely smooth, but has roughness at the atomic level, such as steps, corners, kinks, vacancies, and adatoms, which are extraordinarily more active as bond-breaking catalysts than the smooth level regions. Somorjai showed how to make a surface with a large fraction of atomic scale steps by cutting a single crystal along high Miller index axes.[2,34] Somorjai determined the structures of molecules adsorbed on metal or oxide surface and eventually produced generalizations about the nature of surface chemical bonds.[5,6] His refinements of low energy electron diffraction (LEED) surface crystallography through the development of tensor LEED calculations has led to the important result that most chemisorbed atoms and molecules restructure the metal surface upon adsorption. This has led to the concept of the flexible surface and a proposed relation between reactivity and restructuring ability.[7]

Somorjai showed that the surface structure of a metal and thereby its catalytic activity can be modified in a controlled manner by addition of such components as alumina or oxygen. Also, he found that the strength of surface−adsorbate bonding, and thereby the catalytic activity, can be controlled in one direction by trace addition of potassium and controlled in the other direction by addition of sulfur.[8−11] He found that certain bimetallic interfaces, such as Pt−Au or Pt−Re, and certain metal−metal oxide interfaces have extremely high catalytic activity. The effect of surface structure on catalytic processes were intensively studied on single crystals using a few model reactions, involving hydrocarbons, ammonia synthesis, and hydrogenation reactions. Somorjai found catalytic metal surfaces become coated while in use with a monolayer of carbon atoms or sulfur atoms; he recently showed that these overlayers modify, but do not eliminate, the catalytic activity of the substrate. He has built complex, multicomponent catalysts using single crystal surfaces with proper surface structures as substrates. Somorjai's practical contribution has been to demonstrate to industrial chemists what type of surface structures they must make in order to obtain a desired catalytic reaction rate and selectivity.[12−17]

He has been a major developer of new instruments, and he adapted many physical methods to the needs of his science.[18−21] Somorjai uniquely developed and used a high pressure−low pressure cell for sequential characterization of an initially clean catalyst surface, carrying out a high pressure catalytic reaction and, after the reaction was complete, restudying the catalyst surface at low pressure.[19] Recent applications of two new techniques, sum-frequency generation (SFG)[21] and scanning tunneling microscopy (STM)[20] are of special note.

In early 2000, Somorjai instituted a new direction in his laboratory in order to continue his pursuit to understand the molecular ingredients of catalytic activity. At this time at Berkeley, numerous groups within the chemistry department were making significant strides on how to synthesize nanostructures with exquisite size- and shape-control in solution using concepts from colloidal and crystallization chemistry. Somorjai noted that these methods could be used to synthesize next generation model catalysts − which are referred to as 3-dimensional model catalysts − since they possessed nanoparticles whose size and/or shape was well-controlled, enabling the statistics of the surface atoms and exposed surfaces to be determined or estimated with higher precision than capable with "conventional" synthesized supported metal catalysts.[22−24] He conducted many studies on size- and shape-controlled nanoparticles, but there were parallel studies conducted on single crystals using high-pressure reactivity measurements coupled with surface analysis by sum-frequency generation spectroscopy. Much of this work represents systematic studies of the influence of particle size on the reaction selectivity.[25−27]

After the establishment of SFG to study the catalytic solid−gas interface utilizing single-crystal substrates, he extended the technique to study "soft" interfaces, both solid−gas and solid−liquid. The primary objective of this research thrust was to understand the role of molecular ordering at various interfaces in order to better understand at the molecular level phenomena such as biocompatibility, adhesion, and friction. Surface studies of polyolefin and other polymer blend surfaces, such as Biospan, constituted the subject of his initial studies in this area.[28−30] In these studies, Somorjai was interested in observing changes in the surface as a function of temperature and blend composition, and as the interface is altered from air (hydrophobic) to water (hydrophilic). Professor Somorjai's work extended into the realm of biointerfacial science with a particular emphasis on protein adsorption onto solid surfaces in the early 2000s. Continuing his multitechnique approach, he has primarily used SFG, atomic force microscopy (AFM), and quartz crystal microbalance (QCM), along with several other techniques, to probe the adsorbed structure of proteins,[31] peptides,[32] and amino acids[33] at various interfaces including model hydrophobic and hydrophilic surfaces, in addition to real biomaterials provided by his collaborators from the Polymer Technology Group. His work has focused on elucidating the chemical nature and molecular structure of adsorbed biomolecules. A quote from the new edition of his textbook, *Introduction to Surface Chemistry and Catalysis* (2nd ed.): "What we really want to know is the following: which protein segments are in contact with the substrate, which would enable us to elucidate how the protein attaches to the substrate, and which protein segments are exposed to the solution, so the further adsorption of proteins or cells...can be predicted...the improvement of available techniques and the development of new techniques should be our main focus in the future."

Finally, we would like to acknowledge his overarching contribution beyond surface science. Professor Somorjai has taken a keen and continuing interest in the lives and careers of his students, postdocs, visiting scientists, and colleagues. He has served not just as a teacher and a scientific example but as a friend and a mentor. His advising style involves nurturing young chemists both as scientists and as human beings. Most (if not all) past members of his group have fond memories of their time in Berkeley and of their interactions with Professor Somorjai. He also taught us the importance of relentlessly pursuing new ideas. He encouraged us to pick a new path when given the choice between old and new.

Seong H. Kim
Guest Editor, Department of Chemical Engineering, The Pennsylvania State University

Fabio H. Ribero
Guest Editor, Department of Chemical Engineering, Purdue University

Robert M. Rioux
Guest Editor, Department of Chemical Engineering, The Pennsylvania State University

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Roughness at the Atomic Level, Steps, Corners, Kinks, Vacancies, and Adatoms

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