The Mechanisms of Heterogeneous Catalysis

November 12-14, 1990

Materials Sciences Division
Center for Advanced Materials
Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

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PROGRAM

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PROGRAM

MONDAY - November 12

8:30 - 9:10 AM	*Catalytic Hydrogenolysis on Metals* **J.H. Sinfelt** - Exxon Research and Engineering Company
9:20 - 10:00 AM	*Simulations and Theoretical Insights Related to Mechanisms of Heterogeneous Catalysis;* **W.A. Goddard III** - California Institute of Technology
10:00 - 10:30 AM	**COFFEE BREAK**
10:30 - 11:10 AM	*Characterization and Reactivity Studies of Platinum/Chromium/H-ZSM-5 Catalysts* **R.W. Joyner** - University of Liverpool
11:20 - 12:00 M	*The Effect of Alloying Palladium with Chromium in Supported Catalysts: Correlation Between the Modifications of the Reactivity and of the Electronic Structure* **A.J. Renouprez** - Institut de Recherches sur la Catalyse
12:00 - 2:00 PM	**LUNCH**
2:00 - 2:40 PM	*Atomic Scale Surface Properties of Platinum-Based Alloys* **J.C. Bertolini** - Institut de Recherche sur la Catalyse
2:50 - 3:30 PM	*A Theoretical Study on the Thioresistant Behavior of Small Mixed Platinum-Palladium Particules* **B. Bigot** - Ecole Normale Supérieure de Lyon and Institut de Recherche sur la Catalyse sur la Catalyse
3:30 - 4:00 PM	**COFFEE BREAK**
4:00 - 4:40 PM	*Coordination and Catalysis of Palladium in Zeolite Cages* **W. Sachtler** - Northwestern University
4:50 - 5:30 PM	*Quantum Chemistry of Zeolite Catalysed Reactions* **R.A. van Santen** - Eindhoven University of Technology
Evening	RECEPTION DINNER

All speaker sessions to be held in the Surface Science and Catalysis Laboratory Building 66 Auditorium

TUESDAY - November 13

8:30 - 9:10 AM	*Should We Worry About the Non-Uniformity of Catalytic Surfaces?* **M. Boudart** - Stanford University
9:20 - 10:00 AM	*Structure and Function at Catalyst Surfaces* **J.B. Pendry** - Imperial College, London
10:00 - 10:30 AM	**COFFEE BREAK**
10:30 - 11:10 AM	*Adsorbate-Induced Restructuring on Metal Single Crystal Surfaces* **D.A. King** - Cambridge University
11:20 - 12:00 M	*A General Mechanism for Heterogeneous Catalysis* **L. Salem** (with J.P. Malrieu)- University de Paris-Sud and CNRS
12:00 - 2:00 PM	**LUNCH**
2:00 - 2:40 PM	*Temporal and Spatial Self-Organisation in Catalysis at Single Crystal Surfaces* **G. Ertl** - Fritz-Haber-Institut der Max-Planck--Gesellschaft
2:50 - 3:30 PM	*Cooperative Dissociative Chemisorption as a Possible Mechanism for Catalytic Activity: Static, Kinetic and Dynamic Considerations* **R.D. Levine** - The Hebrew University of Jerusalem
3:30 - 4:00 PM	**COFFEE BREAK**
4:00 - 4:40 PM	*Dissociation of Hydrogen on Clean and Oxygen Covered Pt(111)* **G. Comsa** - KFA-Jülich
4:50 - 5:30 PM	*Chemical Dyamics at Surfaces* **J.C. Tully** - AT&T Bell Labortories
Evening	RECEPTION BANQUET

WEDNESDAY - November 14

8:30 - 9:10 AM	*The Relationships Between Reaction Mechanism and Kinetics in Heterogeneous Catalysis* **A.T. Bell** - University of California, Berkeley
9:20 - 10:00 AM	*Dynamics of the H_2 Dissociation Process* **J.K. Nørskov** - Technical University of Denmark
10:00 - 10:30 AM	**COFFEE BREAK**
10:30 - 11:10 AM	*Oxide Catalysis* **J. Haber** - Polish Academy of Sciences
11:20 - 12:00 M	*Surface Structure Sensitivity in Partial Oxidation Reactions: An Ab-Initio Study of Local Models of (100) and (110) Surface Sites on Molybdenum Oxide (MoO_3) and Their Interaction with Alkenes* **P. Sautet** - Institut de Recherches sur la Catalyse
12:10 - 12:50 PM	*Directions of Theoretical and Experimental Investigations into the Mechanisms of Heterogeneous Catalysis* **G.A. Somorjai** - University of California

All speaker sessions to be held in the Surface Science and Catalysis Laboratory Building 66 Auditorium

ABSTRACTS

In order of presentation

Catalytic Hydrogenolysis on Metals

J.H. Sinfelt

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Metal catalyzed hydrogenolysis reactions have long been of interest with regard to their kinetic and mechanistic features. They also provide dramatic examples of the specificity of metal surfaces in catalysis. Moreover, their use as sensitive chemical probes of the nature of metal catalyst surfaces has been demonstrated repeatedly. For these reasons, among others, studies of hydrogenolysis reactions have provided useful insight into factors of importance in the general area of catalysis by metals.

Simulations and Theoretical Insights Related to Mechanisms of Heterogeneous Catalysis

W.A. Goddard III

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We will summarize some of the recent advances in theoretical methods for studying surfaces and reactions and will consider reaction mechanisms of several catalytic systems.

The recent advances in the theory include

(a) The Interstitial Electron Model for metallic systems
(b) The Charge Equilibration method for predicting charges in inorganic systems (for molecular dynamic studies)
(c) The Biased/Hessian method for using quantum chemical methods to predict force fields for use in molecular dynamic studies
(d) Generic force fields for inorganic systems.

Applications will include surface structure and energetics by metals, semiconductors, polymers, and ceramics plus mechanistic considerations of catalytic systems.

Characterization and Reactivity Studies of Platinum/Chromium/H-ZSM-5 Catalysts

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and P. Johnston and E.S. Shapiro*

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The effect of adding varying quantities of chromium, as the oxide, to 0.5% platinum/H-ZSM-5 catalysts for the hydrogenolysis of ethane and propane has been studied at 1 bar and 500-750K. When the catalyst is reduced at 823K before use, chromium decreases the hydrogenolysis activity in all cases, but the extent depend in a complex way on the amount of chromium present. AT 0.75% chromium, propane hydrogenolysis is suppressed by an order of magnitude over the whole temperature range, but the effect on ethane hydrogenolysis is much less marked. Reduced chromium/H-ZSM-5 materials show no hydrogenolysis activity.

The catalysts have been characterized; by x-ray photoelectron spectroscopy after reduction, and directly in situ by x-ray absorption spectroscopy. After reduction at 623k, both methods show the presence of trivalent chromium and metallic platinum particles, which EXAFS and transmission electron microscopy indicate have an average diameter ,15Å. On reduction at 823K, XPS suggests that ca 10-15% of the chromium has been reduced to the metallic state, while EXAFS clearly shows the formation of platinum/chromium alloy particles. The chromium content of the alloy reaches a maximum and then declines, as the chromium content of the catalyst is increased from 0.5% to 2.25%. Changes in the chromium near edge structure, (XANES), are also consistent with partial reduction to the metallic state.

We argue that the behavior of the alloy particle is best explained as an electronic interaction between platinum and chromium, rather than through the ensemble concept.

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The Effect of Alloying Palladium with Chromium in Supported Catalysts: Correlation Between the Modifications of the Reactivity and of the Electronic Structure

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Carbon supported paladium-chromium supported bimetallic catalysts have been prepared and studied by several physical methods.

EDX scanning transmission electron microscopy shows that 25-30 A particles are formed, which have a homogeneous composition. EXAFS performed at both Cr and Pd K edges indicate that the formation of a solid solution depends on the reduction temperature: at 670K only 50% of the chromium is reduced, whereas at 870K, it is fully reduced and inserted in the network of palladium.

The observed modifications of the Pd K x-ray absorption edge in the presence of a small amount of Cr(5 at.%) is discussed.

They can be either attributed to a transfer of charge between chromium to the 4d, 5s levels of palladium or to a chromium induced rehybridization between these orbitals. A comparison with similar effects observed at the LIII edge of platinum in alloys with Fe, Co and Ni is also discussed. An XPS experiment was also performed on these catalysts. Compared to pure palladium, the $3d^{5/2}$ core level of this metal in the bimetallic catalyst, exhibits a shift of 1 eV.

These modifications of electronic structure are correlated with a deep modification of the chemical properties: The hydrogen chemisorption is reduced but the selectivity for the partial hydrogenation of dienes is increased. Also the selectivity for the conversion of N-hexane to aromatics is enhanced.

It is concluded that this modification of the electronic structure of the palladium by chromium modifies the relative adsorption constants of the hydrocarbons, which is confirmed by competitive hydrogenation reactions.

Atomic Scale Surface Properties of Platinum-Based Alloys

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A combination of several surface science techniques applied to single crystals has resulted in a better knowledge of the surface composition and structure of Pt-Ni and Pt-Fe alloys. On (111) oriented samples, Pt is found to segregate in both these alloys.

As compared to pure Pt, the chemisorptive properties have been found to be deeply modified due to the segregation. Moreover, the segregated alloy surfaces have better catalytic properties towards hydrogenation reactions of olefins, dienes and α, ß-ethylenic aldehydes.

The results will be discussed in the light of electronic properties of the systems.

A Theoretical Study on the Thioresistant Behavior of Small Mixed Platinum-Palladium Particules

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It has been experimentally observed that, in the hydrogenation catalysis of alkenes, small mixed platinum-palladium particules offer a much better resistance to the poisoning effect of sulfurized species than the pure platinum particules. Moreover, the substitution of palladium to platinum does not reduce substantially the initial catalytic activity of the metal particules.

In order to understand the origin of this property and to forecast the properties of particules of different atomic compositions, calculations on small metal particles without or with hydrogen sulfide molecules adsorbed on them have been performed by using an Extended-Hückel type hamiltonian and the recursion method. The metal particules are made of pure platinum or of a mixing of platinum and palladium atoms with various stereochemistry.

A comparison of the electronic properties of the various metal centers and of the bonding between the metal centers and the sulfide molecules according to the stereochemistry of the particule and of the position of the metal center within the given particule is presented. This comparison shows that the above described qualitative method is able to serve as a useful guideline to distinguish the metal atoms in their hydrogen sulfide adsorption properties as a function of their surrounding and of the atomic composition of the whole particule.

Coordination and Catalysis of Palladium in Zeolite Cages

W. Sachtler

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The coordination chemistry of $Pd(NH_3)_x^{2+}$ ions in zeolite NaY has been identified by UV-VIS diffuse reflectance and EXAFS. Following calcination at 250°C the Pd ions are located in supercages and coordinated to two oxygens and two ammine ligands; at 500°C bare Pd^{2+} ions are present in sodalite cages and hexagonal prisms.

Controlled reduction at low temperature yields isolated Pd atoms in sodalite cages or very small Pd clusters in supercages. Adsorption of CO results in a significantly increased cluster size with a coordination number of Pd (CN=6), consistent with a Pd carbonyl cluster containing 13 Pd atoms and perfectly fitting in the supercages. Over these catalysts CO is hydrogenated to methane; larger Pd clusters convert CO + H_2 to methanol which is, over acid sites, subsequently converted to DME and branched hydrocarbons.

Pd clusters form adducts with zeolite protons, $[Pd_n\text{-}H_z]^{2+}$, which are electron deficient and give rise to catalytic superactivity. The formation of such adducts is favored by a high concentration of divalent ions which preferentially populate sodalite cages and hexagonal prisms, thus preventing the migration of Pd^{2+} ions into these positions. Formation of Pd_n clusters and protons in supercages during reduction entails formation of $[Pd_n\text{-}H_2]^{z+}$ adducts.

Quantum Chemistry of Zeolite Catalysed Reactions

R.A. van Santen

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Zeolites are used as solid acids and metal particle supports. We will present lattice minimization studies of zeolite stability as a function of composition using potentials developed on the basis of ab-initio cluster calculations. Relaxation of the zeolite lattice is found to play a crucial role. The consequences of lattice relaxation for Bronsted acidity will be analyzed with ab-initio calculations on aluminosilicate rings. Adsorption of NH_3 will be analyzed. The reactivity of small metal particles is significantly affected by the close presence of polarizing cations. This will be demonstrated for CO and H_2 adsorption.

Should We Worry About the Non-Uniformity of Catalytic Surfaces?

M. Boudart

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Yes, because the activation energy and the pre-exponential factor of the rate constant of each elementary step vary markedly with surface coverage. It is important to understand and ultimately predict this general though not universal phenomenon for the successful kinetic design of catalytic cycles. Yet, while the problem must be kept in mind, it should not deter us from attempting kinetic modeling, as the assumption of uniform surfaces leads to acceptable results in the majority of cases. Attempts at explaining this contradiction between theory and practice will be discussed.

Structure and Function at Catalyst Surfaces

J.B. Pendry

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Recent developments in surface crystallography reveal that catalysts are far from being structurally passive substrates for reactants. Adsorption of atoms, such as oxygen, nitrogen, or carbon, has been found to evoke a structural response in the substrate in addition to the well known electronic response.

I shall describe some of the techniques, diffuse LEED, direct methods, etc., which have yielded this detailed structural information, and indicate the advances we can expect in the future. Some specific structures will be shown and their possible consequences for catalytic activity indicated.

Finally, speculations will be made on how we might move forward from studies of the static structure of reactants before and after a reaction at a surface, to probing details of the dynamics. This is an ambitious goal, but one which may be possible for simple systems.

Adsorbate-Induced Restructuring on Metal Single Crystal Surfaces

D.A. King

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Structural studies have revealed cases where both atomic and molecular adsorbates cause a restructuring of the metal surface. A review will be presented of work on W(100), which undergoes a clean surface reconstruction and is also restructured by H, N, O and Cu adatoms, and on the (100) surfaces of the fcc metals Cu, Ag and Pd, which are restructured by alkali metal adatoms and by carbon monoxide (at intermediate coverages).

A General Mechanism for Heterogeneous Catalysis

L. Salem

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and

Jean-Paul Malrieu

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A novel mechanism is presented for heterogeneous catalysis, whereby adsorbate dissociation occurs without activation energy through self-induced metal-mediated ionic enhancement and stabilization of the adsorbate bond.

Temporal and Spatial Self-Organization in Catalysis at Single Crystal Surfaces

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The elementary steps underlying the mechanism of a catalytic reaction are also responsible for its rate under steady-state conditions. Under certain conditions the rate will, however, no longer be stationary, but may become oscillatory or even chaotic. These effects are associated with the formation of spatio-temporal concentration patterns. The theoretical basis has to be sought in the nonlinear character of the underlying coupled differential equations as will be demonstrated for the oxidation of CO on Pt(110), for which system also the corresponding experimental evidence will be presented.

Cooperative Dissociative Chemisorption as a Possible Mechanism for Catalytic Activity: Static, Kinetic and Dynamic Considerations

R.D. Levine

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The Kinetic model for catalytic reactions involving metal surface restructuring [Levine and Somorjai, Surf. Sci. (1990)] is discussed. Particular attention is given to possible experimental tests of the model involving static, kinetic and dynamic experiments.

Dissociation of Hydrogen on Clean and Oxygen Covered Pt(111)

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The dissociative adsorption of hydrogen has been investigated by combining the manifold capabilities of thermal energy atom scattering (TEAS) and of molecular beam relaxation spectroscopy (MBRS): sensitive coverage monitoring; step density and overlayer structure characterization; angular, velocity and reaction-time resolved reaction product analysis. The results emphasize the strong surface structure sensitivity of the dissociative reaction. Mainly two significant aspects will be discussed: 1) The new data lead to a two channel adsorption model, implying that below 200K there is no hydrogen dissociation on the clean Pt(111)-terraces. The model also explains previous data including features which were not understood so far. 2) The presence of disordered adsorbed oxygen reduces the hydrogen dissociative adsorption probability. A strong correlation between the ordering state of the oxygen overlayer and the hydrogen sticking probability via one of the adsorption channels has been uncovered.

The reaction between hydrogen and oxygen to water is extremely sensitive to the state of the surface: on surfaces prepared slightly differently, but appearing identical when analyzed with sensitive methods, the reaction rate differs significantly. Thus, the extraction of general conclusions is at present hardly possible.

Chemical Dynamics at Surfaces

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Elementary chemical events at surfaces are being examined with ever increasing sophistication, utilizing sensitive surface structural and spectroscopic probes, molecular beam control of reactants, and laser quantum state specificity. Dynamical theories are now being challenged by hard data, and current theories frequently fall short. Of particular concern, especially when the surface is metallic, is the ubiquitous assumption that atomic motion is governed by a single adiabatic potential energy surface. Progress toward a unified theory of chemical dynamics at surfaces will be outlined. Examples involving rotational and vibrational energy exchange, and trapping-desorption will be discused, with direct comparison to experiment.

The Relationships Between Reaction Mechanism and Kinetics in Heterogeneous Catalysis

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Accurate description of the kinetics of catalyzed reactions requires knowledge of the reaction mechanism and a theoretical model of the dynamics of each elementary step. The mechanism, or sequence of steps, by which reactants are transformed into products is postulated on the basis of known chemistry and spectroscopic evidence of reaction intermediates. The dynamics of each elementary step are governed by the potential describing the interactions of an adsorbate with the surface. Because adsorbate-adsorbate interactions can influence the energetics of elementary processes, these effects must also be taken into account. This talk will review the current understanding of the dynamics of elementary processes and will illustrate the use of this understanding to develop accurate descriptions of reaction kinetics. Particular attention will be given to the use of stochastic, as opposed to continuum, representations of reaction dynamics, since such representations can take into account the effects of local configurations of adsorbates. Examples of the use of continuum and stochastic models for the description of CO oxidation, the reaction of H_2 and O_2, and NO reduction will be discussed.

Dynamics of the H_2 Dissociation Process

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and

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The dynamics of the H_2 dissociation process is studied by quantum simulations of the motion in a potential calculated within the effective medium theory. The role of tunneling, vibrational and rotational excitation of the molecule, angle of incidence, and the geometry of the surface is investigated. We also contrast adsorption on Cu surfaces where the adsorption is activated with the adsorption on Ni surfaces where the activation energy is small or absent.

Oxide Catalysis

J. Haber

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See addendum

Surface Structure Sensitivity in Partial Oxidation Reactions: An Ab-Initio Study of Local Models of (100) and (110) Surface Sites on Molybdenum Oxide (MoO_3) and Their Interaction with Alkenes

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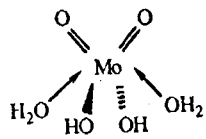
The concept of surface structure sensitive reactions, first introduced for catalysis on metals has more recently been extended to heterogeneous reactions on metal oxides. Within the numerous examples of structure sensitive partial oxidation reactions, the oxidation of propene on molybdenum oxide MoO_3 is one of the best documented cases. The selectivity in acroleine formation with respect to the complete oxidation in CO_2 has been correlated to the ratio between the (100) and (110) exposed faces in the sample, the (100) faces being selective in acroleine production.

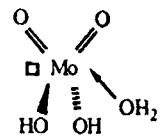
However, important controversies still exist in the interpretation of this data, especially for the specific roles of each crystal face in the different steps of the partial oxidation process.

In this communication we present an ab-initio study of the electronic structure of molecular models for (100) and (110) surface sites of MoO_3. These local models are built from an analysis of the MoO_3 bulk crystal structure that leads to a partition of the M-O bonds in double, single, and dative bonds. This partition results in a simple Lewis electronic structure interpretation of the oxide and is our guide for the extraction of the model molecular sites.

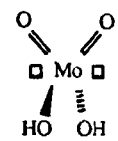
In the perfect bulk, and also in the (110) surface (I), each molybdenum atom is involved in two bonds of each type, while in the (100) face (II) one dative bond is broken leading to the creation of a Lewis acid site on molybdenum. The single bonds arise from bridging oxygens which have been modelled by OH groups while the dative bonds with neighboring Mo-O-Mo fragments are represented by OH_2 molecules.



(I) bulk and (110)



(II) (100)



(III) basic M_o^{VI} unit

The geometry of each one-molybdenum model has been fully optimized. In the case (I), this geometry is compared with the experimental surrounding of molydbenum while for the model (II) special attention is given to the possible occlusion of the unsaturated site by relaxation of the doubly bonded oxygens.

The Lewis acidity on the Mo atom of each model is then compared by interaction with water and ethylene molecules. The adsorption of ethylene on the (110) and (100) sites is studied as a model of the first step of propene activation on MoO_3.

In order to improve the description of the active site, two-molybdenum surface sites are then built from the previous units and partially reoptimized. The interaction of these two-molybdenum models with a propene molecule will finally be described.

Directions of Theoretical and Experimental Investigations into the Mechanisms of Heterogeneous Catalysis

G.A. Somorjai

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The roles of the atomic structure and the electronic structure of the active surface sites in bonding of reactants and causing bond breaking or bond formation have been the focus of theoretical studies. In addition to calculations on static systems, usually clusters, modelling of the transition states and the dynamics of elementary reaction steps (adsorption, dissociation, surface diffusion, desorption) have been performed. Variations of electronic structure of elements across the periodic table have been shown to be responsible for the unique importance of transition metals in catalysis.

Experimental studies utilize catalysts with well-characterized structure (zeolites, crystal surfaces) and information about surface structure, composition and chemical bonding of adsorbates becomes available on the molecular level. Deliberate alteration of catalyst structure, surface composition by alloying and electronic structure by addition of electron donor and electron acceptor promoters have been utilized to modify reaction rates and selectivity. This way many of the molecular ingredients of heterogeneous catalytic reactions have been identified.

In recent years evidence has been accumulating that indicates periodic and long term restructuring of the catalyst surface as necessary for chemical change and reaction turnover. These findings point to the need of time resolved studies and in-situ investigations of both the substrate and the adsorbate sides of the surface chemical bonds simultaneously on a time scale shorter than the reaction turnover frequency.

Close collaboration between theorists and experimentalists is essential if we are to succeed in designing heterogeneous catalysts.

ATTENDEES

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