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PROGRESS REPORT

High Pressure Heterogeneous Catalysis in a Low Pressure UHV Environment

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S. T. Ceyer

Department of Chemistry

Massachusetts Institute of Technology

Cambridge, MA 02139

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The major thrust of our research is to carry out for the first time a heterogeneous catalytic reaction that normally is observed only at high pressures (>1 atm) of reactant gas at low pressures (< 10^{-4} Torr) in an ultrahigh vacuum environment. The reaction we have chosen is the steam reforming of methane on a Ni(111) crystal.

$$CH_4 + H_2O \xrightarrow[Ni(111)]{} CO + 3H_2$$

The problem is that there is a barrier to the dissociative chemisorption of both reactants which is surmountable only by their incident energy. At high pressures, the concentration of high energy CH_4 and H_2O molecules is large enough so that the rate of this reaction is measurable. At low pressures, this concentration is too small for the rate of this reaction to be measurable so the dissociative chemisorption of CH_4 and H_2O must be activated by raising their translational energies. The translational energies of CH_4 and H_2O can be raised through the use of molecular beam techniques. This requires us to build an apparatus which has two molecular beams.

During this past year we have been working on the design and construction of this apparatus. It combines molecular beam surface scattering techniques with two kinds of vibrational spectroscopy, making it a uniquely versatile tool for the characterization of

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surface reactions. Two molecular beams are aimed at the single crystal surface. One of the molecular beam sources is rotatable. This feature, along with the ability to rotate the crystal, will allow us to measure the angular distribution of the scattered products. The scattered reaction products are detected by a triply differentially pumped quadrupole mass spectrometer equipped with a 30 cm flight path for time-of-flight measurements. Time-of-flight measurements allow us to determine the energy of the scattered products. The apparatus has been designed to include not only high resolution electron energy loss spectroscopy (HREELS), which will be used to identify the surface-bound intermediates of the reaction, but also FTIR reflection-absorption spectroscopy. The strict normal dipole selection rule operative with photon excitation will allow the unambiguous determination of the symmetry and therefore the site and geometry of the adsorbed intermediate. However, the strict selection rule also precludes the observation of most of the vibrational modes. Therefore, in order to minimize the chance of misidentification, FTIR must be coupled with HREELS. All vibrational modes are observed with HREELS because selection rules break down under electron excitation. This coupling of photon and electron vibrational spectroscopies is the first of its kind and will constitute a powerful approach to the identification of complex adsorbates and surface reaction intermediates synthesized by molecular beams.

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