II-E Surface Chemical Reactions Studied by NEXAFS Spectroscopy

Near edge X-ray absorption fine structure (NEXAFS) is a very promising method to get the quantitative information of surface adsorbed species. Recently, we have developed an energy dispersive NEXAFS method, which is a technique to measure a NEXAFS spectrum in a certain energy range simultaneously. The Hettrick mount monochromator can be used as a polychromator by fully opening the exit slit. Energy dispersed X-rays from the polychromator irradiate the sample surface. Each position of the irradiated surface emits Auger electrons, of which number is proportional to the X-ray absorption coefficient at the corresponding photon energy. By using a position sensitive electron energy analyzer, we can obtain a NEXAFS spectrum from a monolayer-covered surface with a reasonably high signal-to-noise ratio in several tens seconds. This technique has been applied to the study of surface chemical reactions related to fundamental catalytic reactions.

II-E-1 Direct Spectroscopic Observations of the Water Formation Reaction on Pt(111) by NEXAFS and the Simulations with the Kinetic Monte Carlo and the Reaction Diffusion Methods

NAGASAKA, Masanari[1]; KONDOH Hiroshi[1]; AMEMIYA, Kenta[1]; NAMBU, Akira[1]; NAKAI, Ikuyo[1]; SHIMADA, Tohru[1]; OHTA, Toshiaki[2]
([1]Univ. Tokyo; [2]Univ. Tokyo and IMS)

[*J. Chem. Phys.* **119**, 9233 (2003)]

The catalytic water formation reaction was investigated by the energy dispersive NEXAFS spectroscopy. Oxygen covered Pt(111) surface with a (2×2) structure was exposed to gaseous hydrogen (5.0×10^{-9} Torr) at constant surface temperatures (120–140 K). O *K*-edge NEXAFS spectra were measured during the reaction with a time interval of 35 s. From quantitative analyses for the spectra, the coverage changes of the adsorbed species (O, OH, and H_2O) were obtained at 120, 130 and 140 K, as shown in Figure 1. The reaction is composed of three steps, which are characterized by the induction period (I), fast increase in coverage of OH and H_2O with consuming oxygen (II), and slow conversion of OH to H_2O after the consumption of oxygen (III). It was also found that the maximum OH coverage becomes smaller at a higher temperature. The kinetic Monte Carlo simulation has reproduced the three characteristic reaction steps, from which the steps are explained as follows; in the first step OH domains are created through two-dimensional aggregation of H_2O (I), after the nucleation process the second step sets in where the OH domains propagate by the auto-catalytic cycle until they contact with each other (II), and finally the merged OH domains convert to H_2O (III). The Reaction Diffusion simulation has revealed that the density of H_2O nuclei decreases due to its high diffusion rate at a high temperature, which results in reduction of the OH coverage.

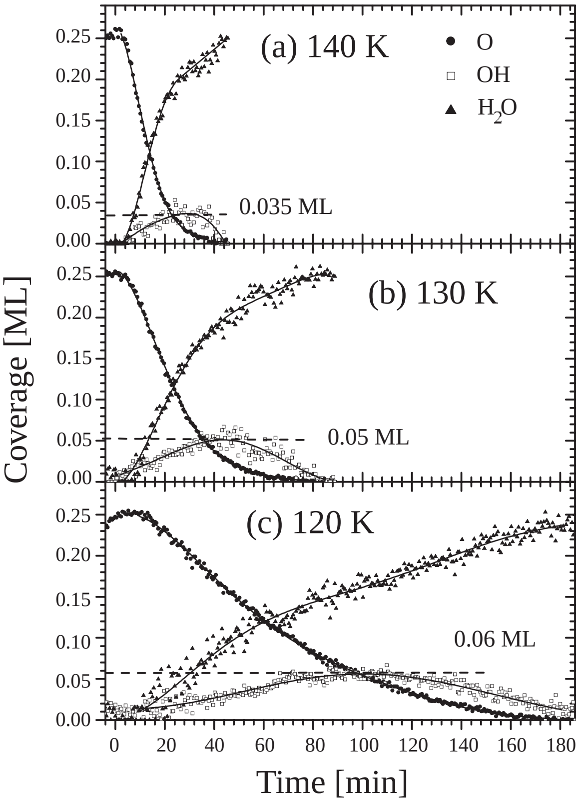

Figure 1. Coverages of O, OH, and H_2O as a function of time during dosing of gaseous H_2 (5.0×10^{-9} Torr) at a surface temperature of (a) 140 K, (b) 130 K, and (c) 120 K. The O-covered Pt(111) surface started exposing to gaseous H_2 at 0 min. The maximum OH coverage was 0.035 ML at 140 K, 0.05 ML at 130 K, and 0.06 ML at 120 K. The reaction finished in 45 minutes at 140 K, 80 minutes at 130 K, and 180 minutes at 120 K.