

O₂ pressure dependence of initial oxidation kinetics on Ni(111) surfaces

Shuichi Ogawa¹, Ryo Taga¹, Tsukasa Ozaki¹, Akitaka Yoshigoe², Yuji Takakuwa¹

¹IMRAM, Tohoku University, 2-1-1 Katahira, Aoba-ku Sendai, Japan

² Japan Atomic Energy Agency, 1-1-1 Koto, Sayo-cyo, Hyogo 679-5148, Japan
ogasyu@tohoku.ac.jp

Nickel (Ni) is used as a catalyst for ammonia decomposition and nitrogen oxide reduction. On the other hand, these catalytic abilities are lost when the Ni surface is oxidized. Therefore, for high efficiency Ni catalyst, it is necessary to prevent Ni surface from being oxidized or to reduce the Ni oxide. Research on the Ni surface oxidation kinetics is indispensable for the development of these catalyst technologies. In this study, O₂ pressure dependence of the initial oxidation kinetics on Ni (111) surface was investigated. Real-time photoemission spectroscopy using laboratory ultraviolet light source and high brightness synchrotron radiation reveals the oxidation mechanism on the Ni (111) surface from oxidation rate, electric structure, work function, and chemical bonding state.

UPS and XPS using synchrotron radiation measurements were conducted at Tohoku University and BL23SU at SPring-8, Japan. The Ni(111) surface was cleaned by repeating Ar⁺ ion sputtering and annealing at 750°C in ultrahigh vacuum. After the start of photoelectron spectroscopy measurements, O₂ gas was introduced directly into the measurement chamber to oxidize the Ni(111) surface. During the oxidation reaction on the Ni surface, the photoelectron spectra were repeatedly measured, and the time evolution of the spectra was obtained. In UPS measurements, secondary electron cutoff and O 2p peak were measured, and work function and the amount of adsorbed oxygen were obtained from these spectra. The O₂ pressure was varied from 1×10⁻⁶ Pa to 2×10⁻⁵ Pa to investigate the O₂ pressure dependence of the oxidation kinetics.

Figure. 1 shows the O₂ molecular dosage dependence of the amount of adsorbed oxygen and work function. When comparing the O₂ molecular dosage (1 L = 1.33×10⁻⁴ Pa·s), the oxygen uptake curve was almost same among all pressures. The amount of adsorbed oxygen sharply increased up to 10 L, and the increment became gentle from 10 L to 20 L. After that, it increased rapidly again after 20 L. And then, the oxidation reaction seemed to stop at 80 L. As described above, it was revealed that the oxygen adsorption on the Ni(111) surface does not depend on O₂ pressure. On the other hand, the work function showed the significant pressure dependence. The work function increases until 10 L after oxidation starts but then decreases. There was no pressure dependence up to 20 L, but after 50 L, the rate of work function decrease became faster with the decrease of O₂ pressure. This result suggests that the lower the O₂ pressure, the more diffusion of O atoms into Ni substrate. In the conference we also show the results of the oxidation states analysis by XPS.

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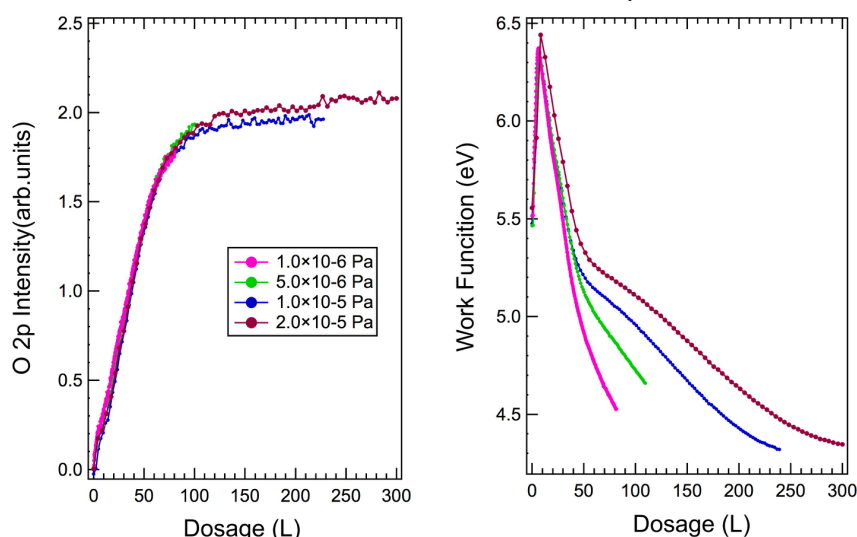


Fig.1 O₂ exposure dependence of O 2p intensity and work function