CO oxidation on Ir(111) surface studied by dispersive-NEXAFS

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Introduction

Platinum group metals have been widely used as catalysts that remove nitrogen oxides (NO_x) and the carbon monoxide in automobile exhaust. It is reported that iridium metal shows the high efficiency in the selective reduction of NO with CO under the oxygen atmosphere. The origin of the high efficiency has not been revealed yet, and it is important to clarify the mechanism of the reaction in order to improve the efficiency further. Here, we have studied CO oxidation reaction (O_{ad} + CO_{ad} -> CO_{2g}) on Ir(111) surface by dispersive near edge x-ray absorption fine structure (dispersive-NEXAFS) spectroscopy.

Experimental

The experiments were carried out at BL-16A_2 of Photon Factory. First, an atomic-oxygen covered surface was prepared on a clean Ir(111) surface. The surface was then exposed to gaseous CO (5.0×10^{-8}–4.0×10^{-7} Torr) at a constant temperature (350~550 K). Dispersive-NEXAFS spectra were taken every ~30 msec under the progress of the reaction.

Results and Discussion

Fig. 1 shows the dispersive-NEXAFS spectra measured during CO oxidation reaction on Ir(111). The rate constants were obtained by analyzing the time dependence of CO and atomic O coverages. Fig. 2 shows the Arrhenius plot, indicating that the frequency factors and the activation energies are different between high and low temperature regions (Table 1), and rate constants of CO oxidation reaction depend on CO pressure only in the high temperature region. In addition, the CO adsorption curves exhibit some flexional points at the boundary temperatures between the high and low temperature regions (Fig. 3). It is likely that the adsorbed oxygen atoms undergo a phase transition at a certain oxygen coverage, and the reaction path would be switched over at the transition.

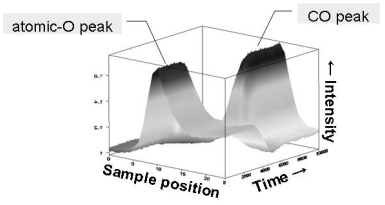

Fig. 1 Dispersive NEXAFS spectra at 425 K under 5×10^{-8} Torr CO gas.

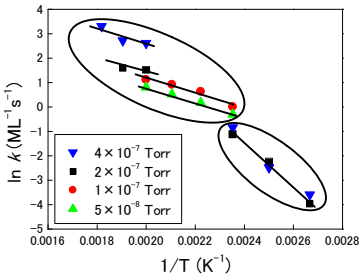

Fig. 2 Arrhenius Plot.

	A_0 / $ML^{-1}s^{-1}$	E_a / eV
Ir(111) at high temperature	$10^{3.1\pm0.4}$	0.27±0.02
Ir(111) at low temperature	$10^{8.7\pm0.7}$	0.77±0.05
Pt (111) [a]	$10^{8.9\pm2.1}$	0.62±0.14

Table.1 Comparison of frequency factor and activation energy between high and low temperature regions. The results for Pt (111) are shown as reference.

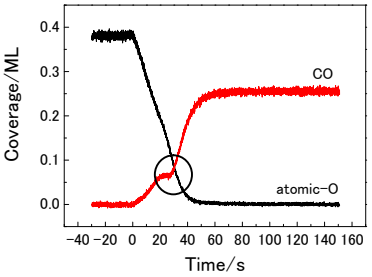

Fig. 3 The flexion point of CO adsorption curve at a boundary temperature (450 K).

Reference

a) Y. Kousa et al, *90th Annual Meeting of The Chemical Society of Japan* **2009**, 2E1-05.