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NO reduction by CO on Ir(111) surface under near ambient pressure conditions

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1 Introduction

Automobile exhaust contains harmful gas such as carbon monoxide (CO), nitric oxide (NO), and so on. Platinum Group Metals (PGM) have a good catalytic performance for conversion of automobile exhaust to harmless gas (i.e. CO oxidation, NO reduction).

Iridium (Ir), one of the PGMs, exhibits a high NO conversion and nitrogen (N_2) selectivity. In this study, NO reduction by CO on Ir(111) surface was investigated in terms of correlation between the catalytic activity and surface chemical states under near ambient pressure conditions using combination of near ambient pressure X-ray photoelectron spectroscopy (NAP-XPS) and quadrupole mass spectrometry (QMS).

2 Experiment

NAP-XPS and QMS measurements were performed at BL-13B of KEK-PF. The Ir(111) surface was cleaned by repeated cycles of Ar^+ sputtering, annealing and oxygen treatment. Cleanness and ordering were checked by XP spectra and low energy electron diffraction.

Under 50 mTorr NO and 10 mTorr CO conditions, QM spectra and XP spectra were recorded simultaneously while increasing the Ir(111) temperature from 303 K to 793 K. To distinguish N_2 from CO and N_2O from CO_2 on QMS, isotope-labeled CO (^{13}CO) was used. The incident X-ray energies used on XPS measurements were 400, 500 and 650 eV for C 1s, N 1s, and O 1s region, respectively.

3 Results and Discussion

Figure 1 shows the partial pressure of products (N_2 and $^{13}CO_2$) (upper) and the adsorbate coverage (lower). The N_2 and $^{13}CO_2$ partial pressure increased at two different temperature, 580 K and 720 K. At 303 K, no reaction takes place and only CO (CO_{ad}) is adsorbed on the Ir surface. At 530 K, the adsorbed atomic nitrogen (N_{ad}) appeared even though the reaction didn't yet start. It indicates that this reaction is not NO dissociation limit process. The reason why coverage of adsorbed atomic oxygen (O_{ad}) is almost 0 monolayer (ML, 1 ML defined as one adsorbate per Ir surface atom) is thought to be that O_{ad} is removed quickly by CO_{ad} forming $^{13}CO_2$. At 580 K, the first catalysis activation, the coverage of CO_{ad} (θ_{CO}) decreases to about 0.3 ML. Since NO can adsorb at atop site below θ_{CO} of 0.27 ML [1], the reaction pathway at this temperature is considered as $N_{ad} + NO_{atop} \rightarrow N_2 + O_{ad}$. At 720 K, the second catalysis activation occurs, where θ_{CO} decreases further and θ_N increases to 0.28 ML. As increasing temperature up to 793 K, however, θ_N decreases to 0.16 ML. It indicates that N_2 desorption rate is faster than adsorption and dissociation rate of NO. The second reaction pathway is considered as $N_{ad} + N_{ad} \rightarrow N_2$.

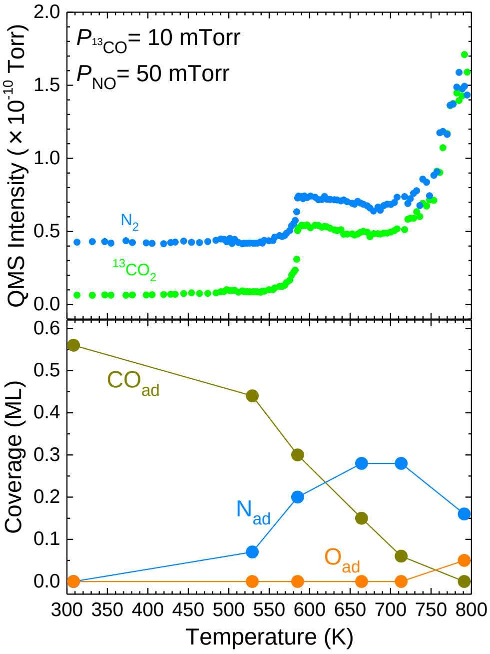

Figure 1: The partial pressure of products (N_2 and $^{13}CO_2$) measured by QMS (upper) and the adsorbate coverage estimated from XPS (lower) versus temperature under NO 50 mTorr and ^{13}CO 10 mTorr.

4 Summary

NO reduction by CO on Ir(111) was investigated by combination of NAP-XPS and QMS. The activation occurred at two different temperatures, 580 K and 720 K. This reaction is not NO dissociation limit process because N_{ad} appeared before the reaction starts at 530 K. At 580 K where the first catalytic reaction starts, θ_{CO} decreases to about 0.3 ML. It suggests that the reaction producing N_2 at this temperature is $N_{ad} + NO_{atop} \rightarrow N_2 + O_{ad}$ because NO can adsorb at atop sites when θ_{CO} is below about 0.3 ML. On the other hand, the reaction producing N_2 at the higher temperature, 720 K, is considered as $N_{ad} + N_{ad} \rightarrow N_2$.

References

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