

Particle Size Dependence for Redox Property of Supported Ni Catalyst Prepared by Impregnation Method

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INTRODUCTION

Under the catalysis reaction environment, the reactant and product gases frequently affect the chemical state of the active metal species. Such chemical state changes give a significant effect on the catalytic performance, and thus the chemical state evaluation is necessary to understand the catalytic performance and provides an essential guideline for the functional catalyst design. The particle size of active metal species is one of the most important parameter for catalytic performance. The purpose of this study is to clarify the chemical state conversion of the Ni species with different particle size under the reactive gas environment.

EXPERIMENTS

The silica-supported Ni particle was prepared by the impregnation method. The stoichiometric amount of organic additives such as citric acid and malonic acid was added into the nickel nitrate aqueous solution to control the Ni particle size.

The XAFS measurements were carried out at BL-9C station of the Photon Factory (KEK, Japan). The *in-situ* XAFS experiments were performed at the Ni K edge during the temperature-programmed oxidation (TPO) and reduction (TPR) processes under the 10 vol.% O₂ or H₂ gas balanced by He, respectively. The results were compared with those for the corresponding Ni catalysts prepared by the standard impregnation method with no additives [1].

RESULTS and DISCUSSION

The particle size of the Ni(0) species was estimated by the TEM observation. The Ni particle size controlled by organic additives was 3 nm (citric acid) and 6 nm (malonic acid).

The particle size was smaller than that of the corresponding Ni catalyst (17 nm) prepared without additives.

The composition changes of the supported Ni(0) species during the TPO and TPR processes are shown in Fig. 1 as a function of temperature. A partial oxidation was observed after the O₂ injection at room temperature for all Ni catalysts. It is considered that the partial oxidation proceeds at the particle surface because the proportion of the oxidized species is almost proportional to the surface area of the supported Ni particle. The oxidation of the Ni(0) species was completed until 300 °C for small Ni particle, which was marked contrast to that of the larger particle, of which the oxidation completed at higher temperature. The opposite changes were observed during the TPR process, and the reduction temperature of the small NiO particle was shifted to higher temperature for the smaller particle. The redox property of supported Ni species is drastically affected by the Ni particles size, and the interaction between the NiO species and the supporting SiO₂ surface contributes to the chemical state distribution.

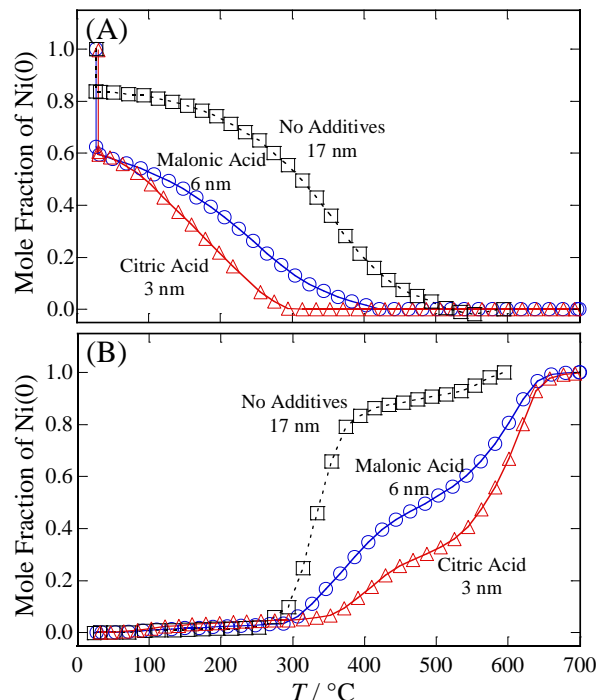


Fig.1 The mole fraction of Ni(0) as a function of temperature during the TPO (A) and TPR (B) processes.

REFERENCES

[1] S. Yamashita, Y. Yamamoto, M. Katayama and Y. Inada, Bull. Chem. Soc. Jpn., 88 (2015) 1629.