Catalytic reduction of NO to ammonia by H_2 or CO-H_2O over metal oxide supported catalyst.

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1.Introduction

Ammonia is an important *N*-containing chemical which is used as fertilizer, starting material for heterocycles sthe synthesis and carbon-free fuel. Ammonia is synthesized industrially by Haber-Bosch method with high energy consumption. Ammonia synthesis from air pollutant NO is an attractive alternative to reduce the concentration of NO. Several reductants such as hydrocarbon, H_2 or CO-H_2O have reported for the transformation of NO to NH_3. Recently, our group developed Pt/TiO_2 catalyst for ammonia synthesis on NO-CO-H_2O reaction.[1] However, the activity of Pt/TiO_2 was not studied for NO-H_2 reaction. In this study, we investigated and optimized the catalytic activity of Pt/TiO_2 for NH_3 synthesis. Furthermore, the activity of Cu/CeO_2 catalyst at lower temperature (>200 °C) investigated for NO-CO-H_2O reaction.

2. Experimental

Supported metal catalysts were prepared by an incipient wetness method with different Pt or Cu precursors. TiO_2 or CeO_2 support was prepared by sol-gel method using different conditions. All catalysts were characterized by BET and CO adsorption. The catalytic activity was measured by a fixed-bed flow reactor. For NO-H_2 reaction, the feed gas was composed of 0.1 % NO, 0.3% H_2 ppm with dilution by Ar. For NO-CO-H_2O reaction, the feed gas was composed of 0.1 % NO, 0.3% CO and 1% H_2O ppm with dilution by Ar. The total flow was set to 250 mL/min. The product gases were analyzed online Fourier transform infrared spectroscopy and gas chromatography.

3. Results and Discusion

Initially, we synthesized Pt/TiO_2 catalyst using different precursor such as $(NH_3)_4Pt(NO_3)_2$, $(NH_3)_4Pt(Cl)_2$, $(NH_3)_4Pt(OH)_2$ and H_2PtCl_6 and tested for NO-H_2 reaction. All precursors showed full conversion (100 %) of NO. The catalytic activity of all four precursors for ammonia selectivity was almost similar at lower temperature (100 °C-150 °C). When the temperature increased to 230 °C, H_2PtCl_4 showed superiority (99 %) for the synthesis of ammonia (Fig.1). Later, we investigated the effect of reduction temperature using different temperatures (400 °C, 500 °C and 600 °C). When Pt/TiO_2 catalyst prepared from H_2PtCl_4 precursor reduced at 400 °C, high yield of ammonia (98.9%) was obtained at 230 °C.

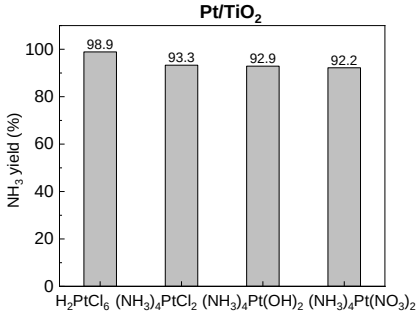

Fig. 1. The activity of Different Pt catalysts at 230 °C

For NO-CO-H_2O reaction, we examined Cu/CeO_2 catalysts prepared from different precursors at 150 °C. The catalyst prepared from $Cu(NO_3)_2$ precursor showed higher yield (72%) than other precursors.

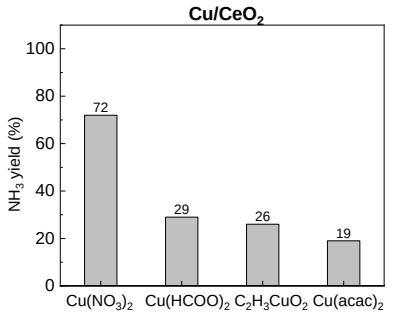

Fig. 2. The activity of Different Pt catalysts at 150 °C

In summary, Pt/TiO_2 prepared from H_2PtCl_6 found to be an active catalyst for ammonia synthesis over NO-H_2 reaction. In NO-CO-H_2O reaction, Cu/CeO_2 catalyst showed high activity for ammonia synthesis.

[1]K. Kobayashi, R. Atsumi, Y. Manaka, H. Matsumoto, T. Nanba, *Catal. Sci. Technol.,* **9**, 289 (2019).