Non-platinum group metal catalysts for ammonia synthesis from NO using CO-H₂O reductant

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Ammonia has a key role in agricultural for production of fertilizers. Traditionally, ammonia is produced by Haber-Bosch method with high energy consumption. Ammonia synthesis from air pollutant such as nitrogen oxide (NO) is an attractive way on the background of NO abatement. Several attempts have been reported for the synthesis of ammonia from NO using Pd, Rh or Pt catalysts. Recently, our group also developed Pt/TiO₂ catalyst for ammonia synthesis on NO-CO-H₂O reaction. However, the cost of Pt group metals is a barrier for their industrial applications. In this study, we have developed the cost-effective Ni/CeO₂ catalyst for ammonia synthesis using NO-CO-H₂O reaction.

CeO₂-supported Ni catalysts were prepared by an incipient wetness method with different Ni precursors. All catalysts were characterized by BET and CO adsorption. The catalytic activity was measured by a fixed-bed flow reactor. For NO-CO-H₂O reaction, the feed gas was composed of 0.1 % NO, 0.3% CO and 1% H₂O 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.

Initially, we synthesized Ni/CeO₂ catalyst using different precursor such as Ni(CH₃CO₂)₂, Ni(NO₃)₂, NiSO₄ and NiCl₂ which were denoted as AC, NO, SO, CL respectively. As shown in figure 1, all catalysts showed high NO conversion above 200 °C except Ni_{CL}/CeO₂ catalyst. Figure 2 shows ammonia yield over different Ni/CeO₂, all catalysts showed low ammonia yield below 200 °C. When the temperature increased to 280 °C, Ni_{AC}/CeO₂

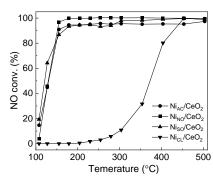


Figure 1 NO conversion over different Ni/CeO₂ catalysts

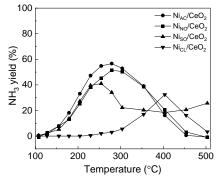


Figure 2 NH₃ formation over different Ni/CeO₂ catalysts

showed superiority (57%) for ammonia synthesis. Ni_{NO}/CeO₂, Ni_{SO}/CeO₂ and Ni_{CL}/CeO₂ catalysts gave 51%, 34% and 3% ammonia yield respectively.

In summary, Ni/CeO₂ prepared from Ni(CH₃CO₂)₂ precursor was an effective catalyst for ammonia synthesis over NO-CO-H₂O reaction.

1) K. Kobayashi, Catal. Sci. Technol. 2019, 9, 2898.