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Full Length Article

Reaction properties of ruthenium over Ru/TiO_2 for selective catalytic oxidation of ammonia to nitrogen

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ABSTRACT

The objective of this study was to investigate reaction characteristics of selective catalytic oxidation of ammonia to nitrogen (NH_3-SCO) using ruthenium-based catalysts. Ruthenium was compared with platinum, a metal generally used as an NH_3 oxidation catalyst. Experiment was carried out by depositing active metal onto TiO_2. 1Ru/TiO_2 was superior to 1Pt/TiO_2 in NH_3 conversion and N_2 yield. Therefore, various analyses were conducted to determine the influence of catalytic properties of 1Ru/TiO_2 on reaction activity. X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS) and Field Emission-Transmission Electron Microscope (FE-TEM) analyses confirmed that ruthenium was present in RuO_2. Such a structure was confirmed to be excellent for adsorption ability of NH_3 and oxygen through analysis of NH_3-TPD, NH_3-TPO, H_2-TPR, and O_2-chemisorption. In addition, in situ diffuse reflectance infrared Fourier transform spectroscopy (DIRFTS) analysis suggested that Ru/TiO_2 had excellent ability to convert NH_3 to NO. The converted NO then reacted with adsorbed NH_3 to generate N_2. Based on results of NH_3 injection at various concentrations, it was found that the rate at which NH_3 was converted to NO from the Ru/TiO_2 surface affected N_2 selectivity.

1. Introduction

Gaseous ammonia is generated from various fixed/mobile pollution sources such as gas slip from selective catalytic reduction (SCR) of NOx using NH_3 or urea in deNOx process, industrial wastewater, chemical process, and semiconductor manufacture process [1]. Ammonia can have serious effects on human health and the environment [2]. For example, ammonia has a serious odor at 50 ppm or less. High concentrations of ammonia can cause serious injuries and burns to the respiratory tract [3]. It has been recently reported that the production of PM2.5 is strongly influenced by the emission of ammonia [4]. Therefore, developing a technology that can control ammonia is becoming very important. There are various techniques for removing ammonia, such as adsorption, absorption, biofiltration, and catalytic oxidation. In the case of adsorption and absorption, operating costs associated with additional processing of adsorbent and absorbent can be significantly high. In the case of biofiltration, it is operated under specific conditions. Various factors such as water content, pH, light, oxygen availability, nutrition, and temperature can have significant impact on its performance. Thus, selective catalytic oxidation of ammonia to nitrogen (NH_3-SCO) method that can convert ammonia selectively to nitrogen and water without additional steps under various conditions is a limelight technology [5]. Various catalysts for NH_3-SCO have been studied. They can be divided into noble metal and transition-metal oxide depending on the active metal. Many transition-metal oxide catalysts such as Fe_2O_3 [6], CuO [7], MoO_3 [8], CoO_3 [9], MnO_2 [10], and CeO_2 [11] have been found to have superior N_2 selectivity. However, high temperatures of 300 °C or higher are required. Therefore, researchers have focused on noble metals such as Pt [12,13], Pd [14], Rh [15], Ru [16], and Ag [17,63] that have excellent activities over a wide temperature range. Among these catalysts, research on Pt-based catalysts having the highest reaction activity has been actively conducted. However, Pt-based catalysts are mainly converted to N_2O which has a problem of low N_2 selectivity [18–20]. Therefore, it is important to develop catalysts with excellent N_2 selectivity at temperature below 300 °C to replace Pt-based catalysts.

Ruthenium is a heterogeneous catalyst with excellent efficiency for CO [21–22], HCl [23–24], and NH_3 [25] oxidation. According to Wang et al. [26], a coordinately unsaturated (cus) Ru atom exposed on the surface of Ruthenium oxide (RuO_2) is excellent for ammonia adsorption and dissociative adsorption of molecular oxygen. Many researchers believe that adsorbed ammonia is converted from the surface of RuO_2

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