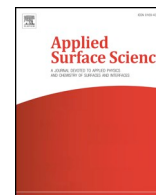




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Reaction properties of ruthenium over Ru/TiO₂ for selective catalytic oxidation of ammonia to nitrogenJung Hun Shin^{a,*}, Geo Jong Kim^{b,*}, Sung Chang Hong^{a,*}^a Department of Environmental Energy Engineering, Graduate School of Kyonggi University, 94-6 San, Iui-dong, Youngtong-ku, Suwon-si, Gyeonggi-do 443-760, Republic of Korea^b University of Kentucky Paducah Extended Campus Program, 209 Crounse Hall, 4810 Alben Barkley Drive, Paducah, KY 42002, USA

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ABSTRACT

The objective of this study was to investigate reaction characteristics of selective catalytic oxidation of ammonia to nitrogen (NH₃-SCO) using ruthenium-based catalysts. Ruthenium was compared with platinum, a metal generally used as an NH₃ oxidation catalyst. Experiment was carried out by depositing active metal onto TiO₂. 1Ru/TiO₂ was superior to 1Pt/TiO₂ in NH₃ conversion and N₂ yield. Therefore, various analyses were conducted to determine the influence of catalytic properties of 1Ru/TiO₂ 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 RuO₂. Such a structure was confirmed to be excellent for adsorption ability of NH₃ and oxygen through analysis of NH₃-TPD, NH₃-TPO, H₂-TPR, and O₂-chemisorption. In addition, in situ diffuse reflectance infrared Fourier transform spectroscopy (DIRFITS) analysis suggested that Ru/TiO₂ had excellent ability to convert NH₃ to NO. The converted NO then reacted with adsorbed NH₃ to generate N₂. Based on results of NH₃ injection at various concentrations, it was found that the rate at which NH₃ was converted to NO from the Ru/TiO₂ surface affected N₂ selectivity.

1. Introduction

Gaseous ammonia is generated from various fixed/mobile pollution sources such as gas slip from selective catalytic reduction (SCR) of NO_x using NH₃ or urea in deNO_x 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 PM_{2.5} 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 (NH₃-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 NH₃-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 Fe₂O₃ [6], CuO [7], MoO₃ [8], CoO₃ [9], MnO₂ [10], and CeO₂ [11] have been found to have superior N₂ 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 N₂O which has a problem of low N₂ selectivity [18–20]. Therefore, it is important to develop catalysts with excellent N₂ 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 NH₃ [25] oxidation. According to Wang et al. [26], a coordinately unsaturated (cus) Ru atom exposed on the surface of Ruthenium oxide (RuO₂) is excellent for ammonia adsorption and dissociative adsorption of molecular oxygen. Many researchers believe that adsorbed ammonia is converted from the surface of RuO₂

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