Applied Surface Science 507 (2020) 145064

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Full Length Article

A study on the structure of tungsten by the addition of ceria: Effect of monomeric structure over W/Ce/TiO_2 catalyst on the SCR reaction

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ARTICLE INFO

Keywords:
W/Ce/Ti
Structural
Monomeric W
Low-temperature SCR
NO adspecies

ABSTRACT

In this study, we investigated the structural characteristics of catalysts and selective catalytic reduction (SCR) reaction activities depending on the Ce valence state and Ce content of the Ce/TiO_2 support in order to examine the influence of the W structural characteristics of W/Ce/TiO_2 catalyst on SCR reaction. Raman, X-ray photoelectron spectroscopy (XPS) and H_2-temperature programmed reduction (H_2-TPR) were performed in order to investigate the physicochemical characteristics of the catalyst·NH_3-temperature programmed desorption (NH_3-TPD) and in situ diffuse reflectance infrared Fourier transform spectroscopy (DRIFTs) experiments were performed to investigate the reaction characteristics of the catalyst. As a result, SCR reaction activity was excellent when the W structure was present as monomeric W, and that this structure was formed by $Ce_2(WO_4)_3$ compound, which was a binding species of WO_4^{2-} monomeric W and Ce^{3+}. In addition, the high Ce^{4+} ratio of the Ce/TiO_2 increased the formation of monomeric W, which was found to be optimal when the loading of Ce was 10 wt%. Finally, the catalyst having the structure of monomeric W showed higher SCR reaction activity at low temperature because of increased NO adsorption and formation of unstable NOx adsorption species.

1. Introduction

In the combustion of fossil fuels of industrial and automobile exhaust gas, nitrogen oxide (NOx) is emitted, which is considered to be a major air pollutant that must be removed [1,2]. NOx can cause acid rain, the greenhouse effect (N_2O), and photochemical smog. The selective catalytic reduction (SCR) of NOx using ammonia is known to be an effective technique for the removal of NOx [3]. It decomposes NOx into harmless gas (N_2, H_2O) through the use of a catalyst in the exhaust gas from various stationary sources [4], as follows:

$$4NO + 4NH_3 + O_2 \rightarrow 4\,N_2 + 6H_2O \quad (1)$$

The SCR catalyst is the most important part of the SCR process for NOx removal, and the V_2O_5-WO_3/TiO_2 catalyst is commonly used. However, V type SCR catalyst has several problems, such as: the volatilization of vanadium, generation of N_2O at high-temperature [5,6], very narrow reaction temperature (300–400 °C) [7], and oxidization of large amounts of SO_2 to SO_3. As the generated SO_3 is reacted with H_2O and NH_3, which is converted to $(NH_4)_2S_2O_7$, NH_4HSO_4, and H_2SO_4, and then it causes the corrosion of the catalyst and plugging of the developing equipment [8]. Recently, due to the various disadvantages of V_2O_5/TiO_2 type SCR catalysts, research on SCR catalysts using new active metals has been conducted.

Tungsten oxide has been extensively studied due to its excellent SCR activity at high temperature range (300–550 °C), such as W/TiO_2 [9]. In addition, cerium oxide has attracted much attention due to its excellent SCR activity at a low temperature range (250–400 °C), such as CeO_2/TiO_2 [10,11]. Recently, it has been reported that the use of tungsten oxide and cerium oxide as active materials showed higher reaction activities than W/TiO_2 and Ce/TiO_2 catalysts [12,13].

Chen et al. [14-16] reported that Ce/TiO_2 added with W showed excellent reaction activity due to the strong interaction between W and Ce. Further, they found through DRIFT research that the addition of W induced a change in the reaction mechanism at low temperature, and reported that the E-R mechanism was the main reaction pathway in the temperature range (150–350 °C). Peng et al. [17,18] showed that CeO_2-WO_3 had excellent SCR reaction activity in the temperature range of 200–500 °C, and reported that the commercial V/W/TiO_2 catalyst had a broader activity window than the CeO_2-WO_3 catalyst. Shan et al. [19,20] reported that the Ce-W catalyst prepared through the homogeneous precipitation method had excellent NH_3-SCR reaction activity in the temperature range of 250–450 °C at S.V. 500,000 h^{-1}. They reported that Ce was highly dispersed over the surface of the catalyst due to the addition of W, leading to increased active sites, oxygen

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https://doi.org/10.1016/j.apsusc.2019.145064
Received 3 November 2019; Received in revised form 11 December 2019; Accepted 12 December 2019
Available online 16 December 2019