Applied Catalysis A: General 435–436 (2012) 156–162

Contents lists available at SciVerse ScienceDirect

Applied Catalysis A: General

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

Promotional effect of F-doped V_2O_5–WO_3/TiO_2 catalyst for NH_3-SCR of NO at low-temperature

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ARTICLE INFO

Article history:
Received 5 May 2012
Received in revised form 28 May 2012
Accepted 30 May 2012
Available online 7 June 2012

Keywords:
F-doped V_2O_5–WO_3/TiO_2
SCR
Oxygen vacancies
W^{5+}
Superoxide ions

ABSTRACT

A F-doped V_2O_5–WO_3/TiO_2 has been developed for low-temperature selective catalytic reduction (SCR) of NO with NH_3. The aim of this novel design was to improve the activity of a catalyst with low WO_3 loading. Analysis by PL spectra, XRD, XPS and EPR showed that F doping improved the interaction of WO_3 with TiO_2 by oxygen vacancies to facilitate the formation of W^{5+} that was important to improve the formation of superoxide ions. The experimental results showed that NO conversion could be improved by F doping and $V_1W_3TiF_{1.35}$ showed the highest NO removal efficiency in SCR reaction at low temperatures.

1. Introduction

Nitrogen oxides (NO_X) are very harmful for the ecosystem and humanity. It contributes to photochemical smog, acid rain, ozone depletion and greenhouse effect [1]. The selective catalytic reduction (SCR) of NO_X with NH_3 in the presence of excess oxygen has been proven to be an efficient process for the removal of NO_X from stationary sources [1–3]. Many catalyst systems, such as V_2O_5–WO_3/TiO_2 [4–6], Mn/TiO_2 [7], CeO_2/TiO_2 [8] and many metal composite oxide catalysts [9,10], have also been extensively investigated. Among these catalysts, V_2O_5 + WO_3/TiO_2 has been applied as an industrial catalyst for many years. The industrial operations are carried out at 350–400 °C [11,12]. However, the high concentration of ash (e.g. K_2O, CaO and As_2O_3) in the flue gas reduces its performance and longevity at this temperature [12,13]. In addition, the high temperature may cause side reactions, such as oxidation of NH_3 into NO and formation of N_2O. However, this could be avoided by locating the SCR unit downstream of the precipitator and even downstream of desulfurizer, through the development of a low temperature (<250 °C) SCR.

WO_3 content of industrial catalyst is about 9% (more details were shown in Table S1 of the Supplementary Material). Many researches have reported the effects of WO_3 in V_2O_5–WO_3/TiO_2 are that: (1) WO_3 increases the amounts of Lewis acid over catalyst [14], (2) WO_3 inhibits the initial sintering of TiO_2 [15], (3) WO_3 improves SO_2 resistance [11], (4) the temperature window for SCR reaction is greatly widened [4]. Recently, efforts have been made to study performance of V_2O_5–WO_3/TiO_2 such as 1.7–2.5% V_2O_5 – 8% WO_3/TiO_2 [16,17], Ce-doped 0.1% V_2O_5 – 6% WO_3/TiO_2 [18], 1% V_2O_5 – 9% WO_3/TiO_2 [4], 1% V_2O_5 – 10% WO_3/TiO_2 [5], 3% V_2O_5 – 9% WO_3/TiO_2 [6], However, these catalysts cannot be applied to investigate the WO_3 characterization and reduce the WO_3 loading.

It has been reported [19,20] that the reduced WO_3 can be oxidized by oxygen and oxygen may be reduced to superoxide ions. The superoxide ions over the catalysts are important to enhance the activity of low-temperature SCR of NO with NH_3 [21]. Therefore, the aim of this study is to improve the interaction of WO_3 with TiO_2 by F doping to increase the number of the reduced WO_3 (W^{5+}) and to promote the SCR activity of a catalyst with low WO_3 loading. To the best of our knowledge, F-doped V_2O_5–WO_3/TiO_2 as a catalyst for NO reduction with NH_3 at low-temperature has not yet been studied. In this study, we found that F doping improved the interaction of WO_3 with TiO_2 by oxygen vacancies to facilitate the formation of W^{5+} that was important to improve the formation of superoxide ions. It was responsible for the improvement of the catalyst by F doping. We also investigated the SCR activity of the catalyst at low temperatures when H_2O and SO_2 existing. Therefore the information in this paper would contribute to a better understanding of the low-temperature SCR processes over V_2O_5–WO_3/TiO_2 catalyst.

2. Experimental

2.1. Catalyst preparation

The F-doped V_2O_5–WO_3/TiO_2 catalyst was prepared by a sol–gel method for F-doped titania preparation, followed by impregnation

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0926-860X/$ – see front matter
http://dx.doi.org/10.1016/j.apcata.2012.05.049