Accepted Manuscript

The promoting effect of tantalum and antimony additives on deNO$_x$ performance of $Ce_3Ta_3SbO_x$ for NH_3-SCR reaction and DRIFT studies

Ning Liu, Jingyu Wang, Fengying Wang, Jun Liu

PII: S1002-0721(18)30158-3

DOI: 10.1016/j.jre.2017.12.009

Reference: JRE 133

To appear in: *Journal of Rare Earths*

Received Date: 31 August 2017

Revised Date: 13 December 2017

Accepted Date: 14 December 2017

Please cite this article as: Liu N, Wang J, Wang F, Liu J, The promoting effect of tantalum and antimony additives on deNO$_x$ performance of $Ce_3Ta_3SbO_x$ for NH_3-SCR reaction and DRIFT studies, *Journal of Rare Earths* (2018), doi: 10.1016/j.jre.2017.12.009.

The promoting effect of tantalum and antimony additives on deNO$_x$ performance of $Ce_3Ta_3SbO_x$ for NH_3-SCR reaction and DRIFT studies

Ning LIU[a], Jingyu WANG [a], Fengying WANG[a*], Jun LIU[b]

[a] School of Environment and Safety Engineering , North University of China, Taiyuan 030051, China

[b] Key Laboratory of coal Science and Technology, Ministry of Education and Shanxi Province, Taiyuan University of Technology, Taiyuan 030024, China

Abstract

A superior Ce-Ta-Sb composite oxide catalyst prepared using homogeneous precipitation method exhibited excellent deNO$_x$ efficiency and nearly 100% N_2 selectivity with broad operation temperature window and better resistance to higher space velocity, meanwhile strong resistance to H_2O and SO_2. This catalyst was systematically characterized using XRD, N_2 adsorption, SEM, TEM, XPS, ESR, Raman, H_2-TPR, NH_3-TPD and *in situ* DRIFTS. There exists a synergistic effect between Ce, Ta and Sb species. It is further indicated that the prominent deNO$_x$ performance of the $Ce_3Ta_3SbO_x$ catalyst is attributed to the elevated Ce^{3+} concentrations, abundant active surface oxygen species, as well as surface acidity and reducibility, which is closely linked with the synergistic effect between Ce, Sb and Ta species. Results from DRIFTS reveal that the reaction mechanism of surface-adsorbed NH_3 and NO_x species is linked to temperature, the L-H mechanism mainly occurs at low temperature (<300 ºC), while the E-R mechanism occurs at high temperature(>300 ºC). Overall, these findings indicate that $Ce_3Ta_3SbO_x$ is promising for NO_x practical abatement.

Keywords: Ce-Ta-Sb mixed oxides; Homogeneous precipitation method; NH_3-SCR; Synergistic effect; NO_x adsorbed species; Rare earths

1. Introduction

The selective catalytic reduction (SCR) of NO with NH_3 is an efficient and economical technique for the removal of nitrogen oxides (NO$_x$) which are major environmental pollutants[1-7]. To date, the commercial V_2O_5-WO_3(MoO_3)/TiO_2 catalysts are active within a temperature window ranging from 300 to 400 ºC. Such commercial catalysts are placed upstream of the de-sulfurizer and electrostatic precipitator, where the catalysts suffered from the poisoning effect of SO_2 and dust[8]. In addition, the poor deNO$_x$ efficiency at high temperature limits their development. Therefore, the research and development of other catalysts with high activities is highly desirable.

It has been found that some transition-metal oxides have catalytic activity for the low-temperature NH_3-SCR of NO, among which manganese oxides exhibit the highest catalytic performance[9,10]. Meanwhile, cerium oxides have already been widely used as promising candidates owing to their prominent oxygen storage capacity and redox cycle ($Ce^{4+}\rightarrow Ce^{3+}$) in the NH_3-SCR process[11-13]. Their acid-base properties, low cost and inherently environmentally friendly property also attract much attention[14]. Recently, much research has been focused on the ceria-based NH_3-SCR catalysts, such as Ce -Ti[15,16], Ce-W-Ti[17], Ce-Mo-Ti[18,19], Ce -Mo[20], Ce-Nb[21], Ce -Mn[22–26], Ce -W[27], Ce -Cu[28], and Ce-Zr[29]. Compared with single component CeO_x, these Ce-based composite oxide catalysts exhibit better deNO$_x$ performance, N_2 selectivity, and the specific surface area. Besides that, Sb has been used as a promoter of some NH_3-SCR catalysts due to its better electric conduction. Zhang et al.[30] studied a $Ce_aTa_bO_x$ catalyst using co-precipitation method, and it exhibited excellent catalytic performance and good resistance to SO_2 and H_2O. Liu et al.[31] studied a series of Ce-Sb binary oxide catalysts prepared by the citric acid method for the NH_3-SCR reaction, and proposed that the strong interaction between Sb and Ce species not only enhances the redox property of the catalyst but also increases the surface

Foundation item: Project supported by the Natural Science Foundation of China (11572292).
***Corresponding author**: WANG Fengying (**E-mail**: fengyingywang@126.com; **Tel.:** +86-351-3921560)