

Selective Catalytic Reduction of NO by CO Over Titania Supported Transition Metal Oxide Catalysts at Low Temperature

Pavani M. Sreekanth and Panagiotis (Peter) Smirniotis *

Chemical & Materials Engineering Department

University of Cincinnati, Cincinnati, OH 45221-0012 USA

Panagiotis.Smirniotis@uc.edu

Introduction

Commercial SCR catalysts use V_2O_5/TiO_2 with either WO_3 or MoO_3 [1,2]. Typically these catalysts operate at medium temperatures (300-400 °C). Low-temperature alternatives of these SCR catalysts for NO_x removal offer enormous potential. Earlier, we developed novel TiO_2 supported transition metal oxide catalysts for low temperature SCR of NO with NH_3 as reductant [3,4]. Here, we report the catalytic activity of titania supported transition metal oxide catalysts for low temperature SCR of NO with CO as reductant. The main purpose of the present study is to exploit the use of these novel catalysts for ammonia free SCR reaction.

Materials and Methods

A series of high surface area anatase titania supported transition metal oxide (M = Cr, Mn, Fe, Ni, Cu) catalysts were prepared by wet impregnation method using dilute solutions of metal nitrate precursors. The prepared materials were calcined at 500 °C for 4 h in air atmosphere.

Results and Discussion

XRD patterns of all the samples showed broad diffraction lines due to anatase phase of titania (JCPDS file no. 21-1272). Except Cu/TiO_2 sample, independent lines from crystalline promoters were not observed in the XRD patterns of titania supported catalysts. This indicates that all the promoter atoms are well dispersed on titania, and they are in amorphous or poorly crystalline state. However, very low intensity CuO lines were observed on Cu/TiO_2 sample. The specific surface areas and the total acidity values of the prepared catalysts are presented in table 1. It could be observed from this table that the surface areas of the prepared catalysts were much lower than the pure TiO_2 support.

Temperature programmed reduction (TPR) patterns of the supported TiO_2 catalysts are shown in Figure 1. It could be observed from this figure that the reduction temperatures of all titania supported oxides decreased when compared to pure individual oxides. The reduction profile of pure CuO is characterized by a single reduction peak at 380 °C. In Cu/TiO_2 system the reduction maximum is shifted to low temperatures indicating a strong interaction between titania support and promoted copper oxide. Low temperature peak is due to dispersed CuO and high temperature peak is due to bulk CuO . Ni/TiO_2 showed two reduction peaks. Low temperature peak is due to reduction of NiO to Ni . High temperature peak is due to reduction of Ti^{4+} to Ti^{3+} . Mn/TiO_2 showed three reduction peaks due to consecutive reduction of MnO_2 - Mn_2O_3 - Mn_3O_4 - MnO .

Catalytic results for the SCR of NO with CO at 200 °C over various transition metal oxides supported on TiO_2 are compared in Figure 2. Under identical operating conditions 10 wt. %

manganese supported on TiO_2 showed excellent performance giving more than 90% NO conversion at the space velocity of 50,000 h^{-1} .

Table 1: Physical properties of the catalysts

| S.no | Sample | BET surface area (m^2/g) | Total acidity (ml/g) |
|------|-----------------|------------------------------|--------------------------|
| 1 | TiO_2 | 309 | - |
| 2 | Cr_2O_3/TiO_2 | 190 | 30 |
| 3 | MnO_2/TiO_2 | 187 | 21 |
| 4 | Fe_2O_3/TiO_2 | 199 | 17 |
| 5 | NiO/TiO_2 | 169 | 17 |
| 6 | CuO/TiO_2 | 55 | 11 |

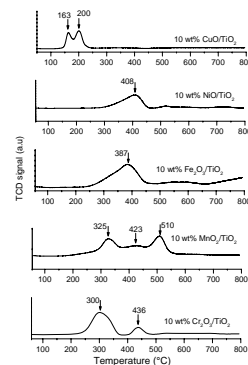


Fig.1 TRR profiles of samples

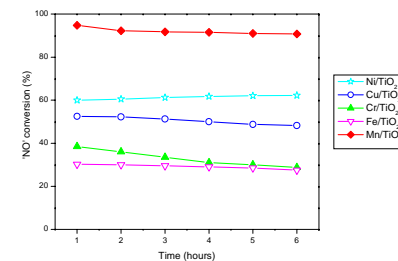


Fig.2 Catalytic activity of samples

Significance

This is a potential alternative for the ammonia SCR process

References

- Dumesic, J. A.; Topøe, N. Y.; Topøe, H.; Slabiak, T, *J. Catal.* **1996**, *163*, 409
- Amiridis, M. D.; Wachs, I. E.; Deo, G.; Jehng, J. M.; Kim, D. S., *J. Catal.* **1996**, *161*, 247
- Smirniotis, P. G.; Peña, D. A.; Uphade, B. S, *Angew. Chem. Int. Ed. Engl.* **2001**, *40*, 2479
- Peña, D. A.; Uphade, B. S.; Smirniotis, P. G, *J. Catal.* **2004**, *221*, 421