REACTIVITY OF NO WITH NH$_3$ IN THE PRESENCE OF O$_2$ OVER Ce-ZSM5 WITH AND WITHOUT MOISTURE

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Abstract
Cerium based ZSM5 catalysts are used to study NO reduction with NH$_3$ in the presence of oxygen with and without moisture. The Ce-ZSM5 was prepared by wet impregnation method and characterized by X-ray diffraction technique, BET surface and SEM. Ce-ZSM5 showed better NO$_x$ reduction than H-ZSM5 which is a poor catalyst for NO$_x$ reduction with NH$_3$. The metal incorporation in H-ZSM5 has increased the catalytic activity. The catalytic activity showed significant difference in NO$_x$ conversion with and without moisture. The disperse Ce species are the active centers for the reduction of NO with NH$_3$ in the presence of oxygen.

Keywords: Catalytic reduction of NO$_x$, ammonia, Ce-ZSM5 with and without moisture

INTRODUCTION

The NO, NO$_2$ and N$_2$O are significant pollutants in the environment discharged by transportation and other stationary sources of pollution. The metal based zeolite catalysts have been shown to be active catalysts for NO$_x$ reduction by ammonia [1-3]. The catalytic reduction of nitric oxide in presence of oxygen is an important process, aiming at cleaning of diesel and lean burnt exhaust gas. The selective catalytic reduction of NO$_x$ with NH$_3$ is the technique applied in stationary sources. Many investigators have attempted NO reduction by CO, H$_2$ and hydrocarbons [4-7], but ammonia type reductants are generally...
more effective for NO\textsubscript{x} reduction than hydrocarbons [8,3]. Ammonia is comparatively less reactive toward oxygen than hydrocarbons [9]. The promising report of Cu-ZSM5 [10,11] for NO\textsubscript{x} reduction has given an impetus to a variety of zeolites to be investigated. In presence of oxygen, NO reacts with NH\textsubscript{3} giving out different products such as N\textsubscript{2}, N\textsubscript{2}O and H\textsubscript{2}O. It is also more important to prevent the formation of N\textsubscript{2}O, which is also one of the significant contributors to the greenhouse effect and the oxidation of ammonia. Catalytic reduction of NO with NH\textsubscript{3} in presence of O\textsubscript{2} seems to be more suitable as far as selective catalytic reduction is concerned. In the present work the activity and selectivity of NO\textsubscript{x} reduction by NH\textsubscript{3} over Ce on H-ZSM5 (designated as Ce-ZSM5) and H-ZSM5 with and without moisture are presented and discussed.

EXPERIMENTAL

H-ZSM5 was supplied by Alsi-Penta Zeolithe; all other chemicals used were from Fluka. The preparation of Ce-ZSM5 was carried out by the wet impregnation method. Appropriately weighed quantity of cerium nitrate was taken with H-ZSM5 (Si/Al = 20) so that the total metal content in ZSM5 was around 3.25 wt.%. Weighed quantity of H-ZSM5 powder was added slowly to the continuously stirred dilute metal nitrate solution. The mixture was then continuously stirred at 343 K for about 15 h on a magnetic stirrer and then heated to dryness. The solid mass was homogenized in an agate mortar and then heated in a furnace at 823 K for 5 h.

The X-ray diffractograms of the samples were recorded on a Siemens Diffractometer (model D-500) using Cu-K\textsubscript{α} radiation filtered through Ni. The total BET surface areas of these samples were measured at liquid nitrogen temperature using Omnisorp (100 CX).

H-ZSM5 was heated to 673 K before use as a catalyst and for SEM micrographs. The morphology of these catalysts was analyzed by SEM on a Cambridge Stereoscan S4 - 10 Microscope. The samples were prepared by spraying catalyst powder in resin melt (Technovit 4071) which then solidified and the sample surface side was polished to smoothness followed by gold metallization in vacuum.

Catalytic activity was measured using a feed gas compositions of 1200 ppm of NO and 1200 ppm of NH\textsubscript{3} in N\textsubscript{2} with 6.5 vol. % O\textsubscript{2}. The same reaction was also studied in moist conditions allowing 5 vol. % water vapor in the reaction mixture. The total Gaseous Hourly Space Velocity (GHSV) was around 180,000 h\textsuperscript{-1}. The reaction was studied in a continuous flow fixed bed quartz reactor in which around 0.2 g of the catalyst powder was loaded in quartz wool.