SCR-NH\textsubscript{3} over V\textsubscript{2}O\textsubscript{5}/Al\textsubscript{2}O\textsubscript{3}·TiO\textsubscript{2} model catalysts: Effect of the alumina content upon activity and thermal stability at high temperature

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Introduction

Acid rain and urban smog, produced mainly by atmospheric pollutants such as NO\textsubscript{x}, SO\textsubscript{2} and volatile organic compounds, are some of the serious global ecological problems [1]. In the case of NO\textsubscript{x} emissions, the Selective Catalytic Reduction (SCR) with NH\textsubscript{3} is probably the best available technology to remove those pollutants from the stack gases of power plants and other stationary sources [1]. Transition metal oxides are the most active SCR catalysts and V\textsubscript{2}O\textsubscript{5}/TiO\textsubscript{2}-based catalysts are the most commonly used around the world. With these catalysts a high NO conversion is attained, close to 98%, between 200 and 450°C. However, above 450°C, the NO conversion decreases progressively [2]. As a result, SCR process requires fine-tuned new catalysts able to remove NO at high temperature.

It has been reported that alumina is one of the most active single metal-oxide catalysts for the SCR of NO with light hydrocarbons at high temperatures, typically above 400°C [3]. However, the studies of the V\textsubscript{2}O\textsubscript{5}/TiO\textsubscript{2}·Al\textsubscript{2}O\textsubscript{3} catalysts have been focused on low temperature (up to 400°C). Here we report the NO reduction with NH\textsubscript{3} under lean conditions over V/Al\textsubscript{2}O\textsubscript{3}·TiO\textsubscript{2} model catalysts at high temperature, above 400°C. We also analyze the effect of the TiO\textsubscript{2}-nanostructures (nano-crystals) as support. The addition of Al\textsubscript{2}O\textsubscript{3} to the support is discussed in terms of the activity and thermal stability of the SCR system.

Materials and Methods

We synthesized several catalysts by the sol-gel method. Titanium isopropoxide and aluminum isopropoxide with 5, 10 and 15 wt. % of Al\textsubscript{2}O\textsubscript{3} were used as precursor for Al\textsubscript{2}O\textsubscript{3}·TiO\textsubscript{2}. The addition of V\textsubscript{2}O\textsubscript{5}, 5 wt. %, to the support was made by wet impregnation. The catalysts were characterized by XRD, FTIR, UV-vis and BET. Samples of 200 mg of catalyst were tested in a tubular quartz reactor mounted in a temperature programmed electric furnace. The reactor temperature was raised from 25°C to 700°C at 4°C/min. The feed stream composition was 500 ppm of NO, 500 ppm of NH\textsubscript{3} and 2% vol of O\textsubscript{2}. N\textsubscript{2} was the gas balance. The NO\textsubscript{x} analysis was made by Chemiluminescence (Rosemount analyzer) in line with an FTIR Spectrophotometer (Bruker Tensor 27) equipped with a 0.75 cm path length infrared gas cell heated at 120°C.

Results and Discussion

The specific surface area (SBET) of our catalysts is close to 330 m\textsuperscript{2}/g. The XRD patterns of the catalysts show the presence of the anatase phase up to 600°C. Rutile was not detected. We found by FTIR that the catalysts present Brønsted and Lewis acid sites. Brønsted acid sites disappear at 200°C. Lewis acid sites remain up to 440°C. In alumina-containing materials NO reduction by NH\textsubscript{3} starts around 160°C over the fresh catalyst, see Fig. 1. NO conversion increases along with the reaction temperature. At 520°C, NO reaches 78% of conversion. NO is hindered between 540°C and 580°C. Above this temperature we found that NO reduction is promoted again.

The addition of 50 ppm of SO\textsubscript{2} to the feed stream seems to promote slightly the NO conversion at low temperature, although the trend closely follows that of the SO\textsubscript{2}-free catalyst. NO\textsubscript{x} reduction in the 550-650°C range is affected, though. This effect depends on the alumina loading, and that will be further discussed, as well as the relationship with surface acidity.

Significance

SCR process requires highly efficient catalysts to remove NO\textsubscript{x} at high temperature to improve the urban air quality. We present here improved catalytic materials focused not only on enhanced activity and selectivity, but on hydrothermal stability and sulphur resistance to comply with environmental regulations.

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Figure 1. NO conversion as a function of the temperature over Ti-15Al-5V model catalyst. 500 ppm of NO, 500 ppm of NH\textsubscript{3}, 2% vol. of O\textsubscript{2}, 50 ppm of SO\textsubscript{2} and 200 mg of catalyst was used.

References