

Soot and hydrocarbon oxidation over vanadia-based SCR catalysts

Enrico Japke, Maria Casapu, Vanessa Trouillet, Olaf Deutschmann, Jan-Dierk Grunwaldt*
Karlsruher Institute of Technology, 76131 Karlsruhe, Germany
*corresponding author: jan-dierk.grunwaldt@kit.edu

Introduction

With the rise in demand of more efficient exhaust gas after-treatment systems due to stricter exhaust gas norms there is a need to further improve the currently applied catalysts or to develop new catalytic systems. This goal has to be achieved without further increasing the overall cost and also the volume of the aftertreatment system. Thus, integration of the CO and HC oxidation and of the NO_x removal functionalities (LNT and/or SCR) into the diesel particulate filter (DPF) design is regarded as a future option for the so called “Four-Way Catalyst”^[1-3]. In this context, elucidating the interaction between the different components and process parameters is of major interest.

Significant increase of the efficiency in treating the polluting gases has been reported for gasoline engines equipped with a TWC coated DPF in addition to the conventional TWC, obtained by distributing the catalyst between the two components^[2]. The ability of the zeolite-based SCR catalysts in oxidizing the soot components has been also reported^[4-5]. With the known potential of vanadium-based catalysts to promote the oxidation of volatile organic compounds^[6-7] the aim of the present study was to provide detailed knowledge on the activity of different V-based SCR catalysts for oxidation of soot and representative PM components. In addition, the impact of HC addition on the SCR of NO_x with NH₃ was studied.

Materials and Methods

The catalysts used in this study were prepared by incipient wetness impregnation of TiO₂ (Anatase, Alfa Aesar) with ammonium tungstate (Fluka) and ammonia metavanadate (Merck). In addition, a fully formulated commercial catalyst has been used as reference. Material characterization has been achieved by XRD, UV-Vis, Raman Spectroscopy, XPS, TEM and N₂-Physisorption.

Activity and selectivity tests were carried out with catalyst coated cordierite honeycombs (400 cpsi). A quartz glass plug-flow reactor has been used and a gas mixtures containing 0-1000 ppm NO_x, 0-1000 ppm NH₃, 0-1000 ppm C₃H₆ and 0-500 ppm C₁₂H₂₆, 10vol.% O₂, 5vol.% H₂O in N₂ was dosed at a GHSV of 50 000 h⁻¹ while monitoring the reaction products with an MKS FTIR instrument. Gas and liquid dosage was controlled by mass flow controllers (Bronkhorst) and the experiments were carried out in a temperature range from 150 to 550 °C.

Soot was deposited on the catalysts with a three cylinders OHC four-stroke diesel engine. The honeycomb was placed in a holder located in the exhaust gas line and has been exposed to defined idle and full duty cycles.

Results and Discussion

The results obtained uncovered for all studied V-catalysts a low selectivity towards CO₂ formation during C₃H₆ oxidation at temperatures higher than 200 °C. Figure 1a shows the oxidation of propylene as a function of temperature over the commercial reference sample with

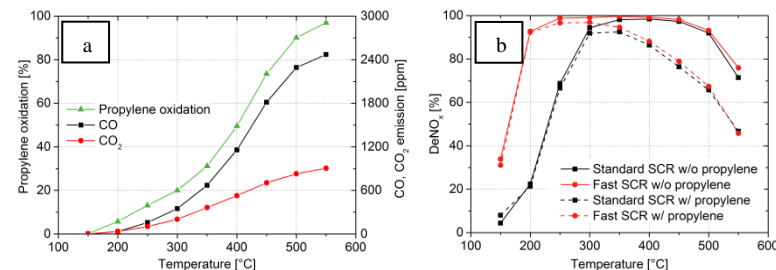


Figure 1. (a) C₃H₆ oxidation over V-SCR. GHSV 50000 h⁻¹ with 1000 ppm C₃H₆, 10vol.% O₂, 5vol.% H₂O in N₂. (b) SCR activity with/without the presence of C₃H₆. GHSV 50000 h⁻¹ with 1000 ppm NO_x, 1000 ppm NH₃, 1000 ppm C₃H₆, 10vol.% O₂, 5vol.% H₂O in N₂.

about 2400 ppm CO and only 750 ppm CO₂ formed at 500 °C. A significant impact of HC presence was measured during the standard and fast SCR activity, which is shown in Figure 1b for the commercial V-catalyst. In the temperature range where the catalyst is able to convert propylene a negative influence on the SCR activity was observed. It seems that in this range the oxidation of propylene is kinetically favored over the reduction of NO_x.

For higher chain HCs such as dodecane a negative influence on the SCR activity was observed over the whole studied temperature window. Apart from the obviously kinetically favored oxidation of dodecane compared to the SCR reaction at higher temperatures also the low-temperature performance seems to be negatively affected.

Significance

The studied V-based SCR catalysts have shown good catalytic activity in oxidizing propylene, dodecane and soot. However, the selectivity towards CO formation was very high. Furthermore, the SCR activity significantly decreased particularly at high temperature and for high HC concentrations.

References

1. B. Opitz, A. Drochner, H. Vogel, M. Votsmeier, *Applied Catalysis B: Environmental* **2014**, 144, 203.
2. S. Spiess, K.-F. Wong, J.-M. Richter, R. Klingmann, *Topics in Catalysis* **2013**, 56, 434.
3. M. V. Twigg, *Catalysis Today* **2011**, 163, 33.
4. F. Schrade, M. Brammer, J. Schaeffner, K. Langeheinecke, L. Kraemer, *SAE International Journal of Engines* **2012**, 5, 958.
5. G. Cavataio, J. R. Warner, J. W. Girard, J. Ura, D. Dobson, C. K. Lambert, *SAE International Journal of Fuels and Lubricants* **2009**, 2, 342.
6. E. Finocchio, M. Baldi, G. Busca, C. Pizarino, G. Romezzano, F. Bregani, G. P. Toledo, *Catalysis Today* **2000**, 59, 261.
7. M. Koebel, M. Elsener, *Industrial & Engineering Chemistry Research* **1998**, 37, 3864.