Multivariate approach for studying accelerated chemical aging of a vanadia-based SCR-catalyst

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Introduction

Vanadia-based catalysts (V_2O_5-WO_3/TiO_2) are being used in diesel engine exhaust treatment for control of nitrogen oxide emissions by NH_3-SCR. A challenge in exhaust treatment is catalyst deactivation by various impurities in the exhaust gas. These poisons could originate from lubrication oil (typcially P, S, Ca, Zn), diesel fuel (mainly S) or engine wear (Fe, Cr, Ni). The introduction of biodiesel gives rise to impurities such as phosphorus, originating from the crop, and alkali metals (Na, K) used as catalysts during production of the fuel.

In this study, the influence of six potentially significant catalyst poisons (Na, K, Mg, P, S, Zn) have been investigated using design of experiments (DoE) for catalyst samples contaminated by wet impregnation. The aim of the study was to determine which single poisons have a significant impact on the V_2O_5-WO_3/TiO_2 catalytic performance and to identify two-factor interaction effects.

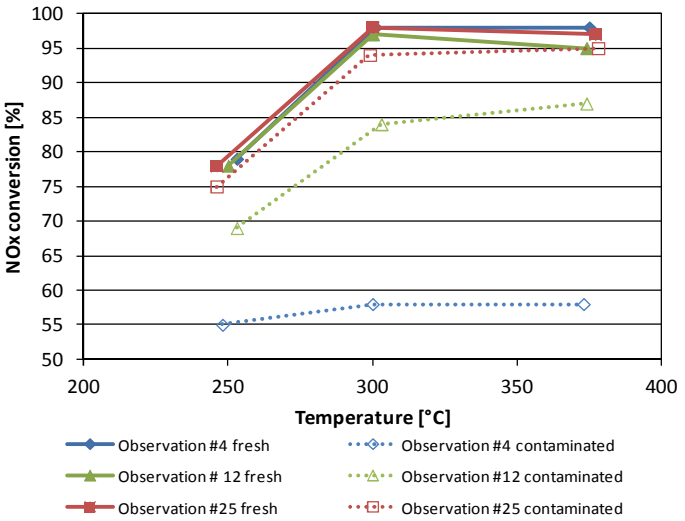
Materials and Methods

The study included:

- Accelerated chemical aging of V_2O_5-WO_3/TiO_2 catalyst samples. A high and a low level of poisoning was included in the experimental design. The target concentrations were 0.25 and 1 wt% for the low and high level, respectively.
- Screening of potentially significant catalyst poisons and possible interaction effects based on a reduced factorial design (2^{6-1}). The performance of the catalysts for NH_3-SCR was measured in a synthetic catalyst activity test rig using the following composition: 1000 ppm NO, 1000 ppm NO_2, 2000/1600 ppm NH_3, 8 % O_2, 6.5 % H_2O and balance N_2. A GHSV of 50 000 h^{-1} was used and the activity was measured at 375, 300 and 250 ºC, respectively, for both fresh and contaminated samples.
- Catalyst characterization by means of ICP, BET, SEM-EDX and NH_3-TPD.
- Multivariate analysis of data
- Comparison with catalysts aged in engine test cell and field test trucks

Results and Discussion

The NO_x conversion for fresh catalyst samples during NH_3-SCR ranged between 75 and 98 %, depending on the temperature studied. The variation between different catalyst samples was low (<1.5 relative standard deviation). Figure 1 shows NO_x conversion for three different observations from the screening study, for fresh and contaminated sample, respectively.



Observation #	P	S	Zn	Na	Mg	K
4	0.25	0.25	0.25	1	1	1
12	0.25	1	0.25	1	1	0.25
25	1	1	0.25	0.25	0.25	1

Figure 1. NOx conversion for three different observations from the screening study. Target concentrations in wt% for each contaminating component in the table below the figure.

The complete set of the experimental design, in total 32 observations, was evaluated by multiple linear regression (MLR). Significant effects were shown for S, Zn, Na, Mg and K as well as for the interactions P·S, P·Na and Na·Mg.

Significance

The study has provided a methodology that can be used to quantify the chemical load of V_2O_5-WO_3/TiO_2 SCR catalysts. The methodology includes tools for accelerated chemical aging in lab scale, as well as deactivation factors, which together with models describing the thermal aging, can be used to predict lifetime of the catalysts.

The results from the study can be used in further and more detailed studies of specific contaminating species that have a negative impact on the catalytic activity and that are likely to exist in future bio-based fuels.