Silver as storage compound for NO_x at low temperatures

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Introduction

The development of engines for a better fuel economy for vehicles has resulted in decreased exhaust gas temperatures, which make it necessary to remove NO_x from the exhaust below 200 °C. The problem with these low temperatures is that the activity for NO_x reduction is very low and it is difficult to dose urea. One solution to this problem is to store the NO_x intermediately in a passive NO_x trap and release it when the principle catalyst has reached operating temperature. For such an application, the material needs to store large amounts of NO_x below 200 °C and easily release NO_x above this temperature to facilitate regeneration. When the NO_x is released from the passive trap it can be reduced over an SCR catalyst, while the trap is regenerated. Another application could be to use the passive NO_x trap before a conventional Pt/Ba/Al containing lean NO_x trap (LNT), where the NO_x can be released at adsorption temperatures in a rich environment and reduced over the Pt/Ba/Al catalyst.

In the present study we show that silver/alumina with a high silver loading (16 wt%) is a suitable material for a passive NO_x trap since it can store large amounts of NO_x when small amounts of hydrogen are present in the feed and release the NO_x below 400 °C in Ar or at adsorption temperature in a rich atmosphere.

Materials and Methods

Silver/alumina catalysts, one with a silver loading of 16 wt% and one additionally doped with 1% of Pt were prepared by incipient wetness. The obtained powders were coated on corderite monoliths and tested in a flow reactor for temperature programmed desorption, cycling experiments with lean and rich cycles and steady state NH_3- and HC-SCR. The same experiments were performed over a conventional Pt/Ba/Al catalyst with 16 wt% BaO and a Pt/Al catalyst.

Results and Discussion

The ability of silver to store NO_x at low temperatures and release it below 400 °C was studied. For this purpose Pt/Ag/Al and Ag/Al with 16 wt% Ag as NO_x storage compound were compared to Pt/Ba/Al (16 wt% BaO) and Pt/Al in the presence and absence of hydrogen. In experiments with periodic rich and lean cycles it was shown that the silver containing samples store higher amounts of NO_x in the presence of small amounts of hydrogen than a conventional Pt/Ba/Al between 175 and 200 °C (see Figure 1). Addition of hydrogen during the lean phase substantially increases the amount of stored NO_x over both the Ag-containing and the Pt/Ba/Al sample between 100 and 200 °C. Propene as another reducing agent does not have the same positive effect as H_2, but in contrary competes with NO for the same active sites. The observed higher NO_x storage capacity might even be due to selective catalytic reduction of NO_x (SCR) when adding a reducing agent during the lean phase in lean-rich cycling experiments. However, the reduction of NO_x with H_2 resulting in N_2 was below 15 % at the studied temperature interval; instead N_2O is formed over the Pt/Al and Pt/Ba/Al catalysts.

These results are confirmed in different NO-TPD experiments where is was, moreover, shown that the stored NO_x is practically completely released below 400 °C, mainly in the form of NO_2 from the silver containing catalysts. The release in the form of both NO and NO_2 is interesting since a potential SCR catalyst after this NO_x trap will operate more effective with this mixture of NO_x.

The results in this study clearly show the potential of using silver as NO_x storage material in two different concepts: (i) Lean NO_x traps with first Ag/Al followed by a conventional barium containing LNT catalyst. NO_x is stored at low temperatures over Ag/Al in the presence of small amounts of hydrogen and then released and reduced over the LNT catalyst. At high temperatures, the LNT stores the NO_x. (ii) Passive NO_x storage catalyst containing silver, followed by urea dosing and SCR catalyst. At low temperatures the NO_x is stored on silver in the presence of small amounts of hydrogen and at higher temperatures released and reduced in the following SCR catalyst.

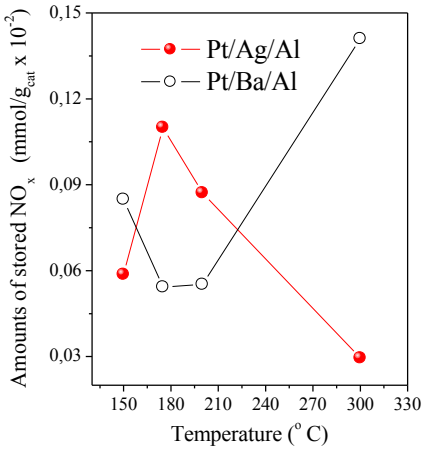

Figure 1. Calculated amounts of stored NO_x species during the lean phase with 1500 ppm H_2 present over the Pt/Ag/Al (red, filled) and Pt/Ba/Al (black, open) as function temperature.

Significance

A high loaded silver/alumina (16 wt%) was found to be suitable as a temporary NO_x trap under cold start, so called passive NO_x storage. High amounts of NO_x can be stored in the presence of H_2 on this material below 200°C and released below 400°C under inert conditions or under rich conditions at adsorption temperature.

References

1. Tamm S.; Andonova S.; Olsson L. *submitted* **2013**.
2. Tamm S.; Andonova S.; Olsson L. *in manuscript* **2014**.