

Low temperature NOx storage using Ag₂O/Al₂O₃ catalysts

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Introduction

Future vehicle emissions standards are getting more and more stringent. California will phase in LEV III emissions regulations beginning in 2015 and the US EPA will implement Tier 3 SULEV emissions regulations in 2017. On the other hand, current internal combustion engines emit the majority of tailpipe exhaust emissions during the first minute or two following "cold-start". This is mainly due to the ineffectiveness of existing catalytic converters below the temperature where they become active [1]. Thus, developing low temperature emissions control technologies has become a top priority of automobile manufacturers and their suppliers. To meet this challenge, unique Ag-based catalysts (Ag₂O/Al₂O₃) were prepared and evaluated as a low temperature NOx adsorber technology. In this paper, we will describe their superior low temperature (< 200°C) NO to NO₂ conversion efficiencies, as well as their low temperature NOx storage capability with the assistance of hydrogen (H₂). Sulfation and desulfation experiments were conducted to examine their sulfur tolerance [2] and benchmark supplier catalysts (diesel oxidation and passive NOx adsorber) were assessed to provide comparisons to the Ag-based catalyst technology.

Materials and Methods

Ag₂O/Al₂O₃ catalysts (1 – 5 wt% Ag₂O) were prepared using the incipient wetness impregnation method. The catalysts were coated onto honeycomb substrate samples to make monolithic catalysts for evaluation using a laboratory fixed-bed flow reactor system. Fourier transform infrared (FT-IR) and mass spectrometers were used for analysis of the gas composition and Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS) measurements were performed to characterize the surface species during NO oxidation. The catalysts were oven aged for 24h at 650°C with 10% H₂O before being evaluated.

Results and Discussion

Compared with Pt/Pd based catalysts, superior NO to NO₂ conversion was observed over the 2 wt% Ag₂O/Al₂O₃ catalyst in the presence of hydrogen < 200°C (Figure 1, left). Unlike Pt/Pd-based oxidation catalysts, the co-existence of light hydrocarbons (propene, propane) had no effect on the NO oxidation activity. The impact of H₂ on NO adsorption was investigated using the in situ DRIFTS technique and the results suggested the formation of nitrates. Both steady state and dynamic NOx storage experiments were carried out and up to 0.19 g NOx per L catalyst was stored below 180°C using 500 ppm H₂ under steady state conditions (Figure 1, right). Furthermore, approximately 50% overall NOx storage efficiency was achieved under a dynamic test condition with periodic NOx and H₂ pulsations at 75, 100, 125, 150, 175 and 200°C. The effect of various operating parameters (space velocity, feed stream composition) on the NOx storage efficiencies and the impact of sulfur poisoning and corresponding catalyst desulfation behavior were also examined. Sulfur poisoning had a detrimental effect on the NOx storage efficiency and desulfation under high temperature rich feed gas conditions was not sufficient to fully recover the initial storage. Additionally, the

technical challenges of the Ag₂O/Al₂O₃ catalysts will be discussed with respect to their NOx storage and release capabilities. Finally, a potential integration method for the Ag₂O/Al₂O₃ catalyst into future diesel aftertreatment system(s) will be proposed.

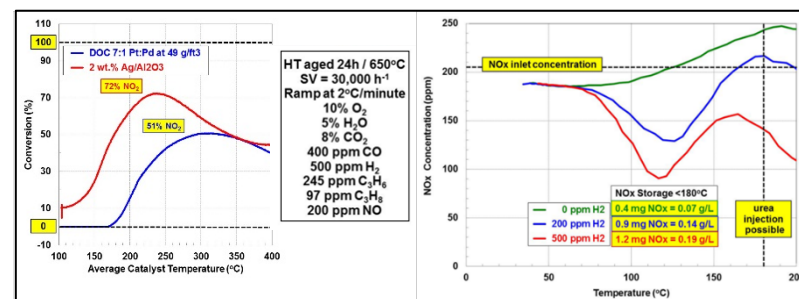


Figure 1. Steady state NO oxidation (left) and NO_x storage (right) over a 2 wt% Ag₂O/Al₂O₃ catalyst.

Significance

Ag₂O/Al₂O₃ catalysts provide an alternative for both low temperature NO oxidation and low temperature NOx storage with the assistance of H₂. This may be imperative for cold-start NOx emissions control when moving towards future SULEV emissions regulations. This work assesses the capabilities of Ag₂O/Al₂O₃ catalysts while providing valuable understanding on the effects of various operating parameters on catalyst performance.

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References

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