INTERPLAY BETWEEN NO_X AND SOOT REMOVAL OVER Pt-Ba NSR CATALYSTS

Nancy Artioli, Roberto Matarrese, Lidia Castoldi, Luca Lietti*, Pio Forzatti

Dipartimento di Energia, Laboratory of Catalysis and Catalytic Processes Politecnico di Milano, P.zza L. da Vinci 32, Milano, Italy

* Corresponding author: Tel.: +39 0223993272, Fax: +39 0270638173, E-mail address: <u>luca.lietti@polimi.it</u>

Introduction

Environmental legislations are imposing strict limits in NO_x and particulate (soot) emissions from Diesel engines. Recently the potential use of NSR catalyst in the simultaneous removal of soot and NO_x has been proposed [1-3]. The aim of the present study is to analyze the effects of the presence of soot on the behaviour of the NSR catalysts. Besides, we want to provide new insights on the role in the soot combustion of NO_x stored onto the catalytic surface.

Experimental

A homemade Pt-Ba/Al₂O₃ (1/20/100 w/w) catalyst was used, prepared by impregnation of γ -alumina with Pt and Ba salts. The effect of the presence of soot on the catalyst activity was analyzed by comparing the behavior of a soot/catalyst mixture with that of a pure catalyst sample. In particular the NO_x storage and reduction activity was studied by performing lean-rich cycles at different temperatures: for this purpose rectangular step feeds of NO (1000 ppm) in He + 3% v/v O₂ and of H₂ (4000 ppm) in He were alternatively fed to the reactor in the presence of CO₂ and H₂O in the feed stream. The soot oxidation during these experiments was also analyzed.

The thermal stability of the stored NO_x in the presence and in the absence of soot was also analyzed by thermal programmed desorption (TPD): after NO_x adsorption at 350°C the sample was cooled in He and then heated up to 500°C. Finally, the behaviour of the catalysts in the soot combustion reaction was investigated by means of temperature-programmed oxidation (TPO) in the presence and in the absence of stored NO_x.

Results and discussion

The results obtained during the lean NO_x adsorption over the Pt-Ba/Al₂O₃-soot mixture showed that the presence of roughly 10% w/w of soot decreases by nearly 30% the NO_x storage capacity of the soot-free catalyst. However the presence of soot does not appreciably affect the behavior of the PtBa/Al₂O₃ catalyst in the reduction by H₂ of the stored nitrates, being in all cases N₂ the major reaction product. The decrease of the NO_x adsorption capability is possibly related to the decrease of the NO₂ concentration, due to its reaction with soot. Besides TPD results pointed out that the thermal stability of the adsorbed nitrates is also decreased in the presence of soot. This indicates an interplay between adsorbed NO_x species and soot: soot affects the NO_x storage but the adsorbed nitrates favor the oxidation of soot, as pointed out by TPO experiments showing that the oxidation of soot is greatly enhanced by the presence of adsorbed NO_x species. These species actively oxidize soot by direct interaction due to their mobility on the surface and/or participate in the soot oxidation upon release of NO₂ and O₂ during decomposition.

Conclusions

This study shows that model Pt-Ba/Al₂O₃ catalysts are able to simultaneously remove soot and NO_x, when operating under cycling conditions, according to the typical NSR strategy. The presence of soot decrease the storage of NO_x and the stability of stored NO_x: this points out that a complex interplay exists between soot and the stored NO_x. Evidences have been pointed out on the participation of the stored NO_x in the soot oxidation, by direct participation of the adsorbed NO_x species in the oxidation of soot and/or upon release of NO₂ and O₂ which actively oxidize soot.

References

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