Revealing the Influence of the Noble Metal-Support Interface on the Structural Dynamics of Pt/CeO₂ by *in situ* Spectroscopy

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Introduction

Avoiding emissions of toxic pollutants like CO and unburnt hydrocarbons (HC) during the cold start of the engine is a major challenge in automotive exhaust gas catalysis. This demands an improvement of the low temperature activity of the diesel oxidation catalysts (DOC) [1]. In this regard, exploiting the interaction between reducible supports and the noble metal component represents a promising approach. Our recent *operando* X-ray absorption spectroscopy results demonstrated that in Pt/CeO₂ catalysts the size of noble metal nanoparticles and consequently the platinum-ceria interface is strongly dynamic and can be tuned by short reducing pulses significantly enhancing the low temperature CO- and HC-oxidation performance [2].

Experimental Method

In this study, a $Pt/CeO_2/Al_2O_3$ catalyst prepared by flame spray pyrolysis was used to study the structural changes of Pt and CeO₂ during temperature programmed reduction and reducing pulses with H₂ and CO at 150 °C and 250 °C. The oxidation state of Pt and Ceria components were monitored by QEXAFS (quick scanning extended X-ray absorption fine structure spectroscopy) at the Pt-L₃ (P64, DESY) and Ce-L₃ edge (SuperXAS, SLS) using a capillary micro reactor [3]. **Results and Discussion**

By using complementary QEXAFS studies at the Pt-L₃ edge and Ce-L₃ edge we were able to elucidate important aspects of the Pt-ceria interface functionality during CO oxidation in a Pt/CeO₂ DOC. The electronic state and the size of Pt particles were shown to be the decisive factor, since reduced Pt nanoparticles are a prerequisite for the reduction of ceria surface sites. The kinetics of ceria reducibility are as well strongly dependent on the Pt particle size. Thus, during the short reducing pulses small Pt entities were formed catalyzing the surface reduction with hydrogen or propylene and increasing the reduction rate of surface ceria and platinum itself. With increasing the number of reductive pulses at 250 °C the noble metal-ceria interface runs through a maximum (highest CeO₂ to Ce₂O₃ reduction extent) due to further Pt sintering, which leads to simultaneous decrease of the ceria reducibility.

Conclusion

The study shows the enormous potential of complementary QEXAFS studies monitoring structural changes of multiple catalyst components and the dynamics of the noble metal-support interaction.

References

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