SINCHEM PhD subject

TITLE: “Simultaneous soot and NOx abatement in light-duty diesel engine vehicle applications”

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PROJECT DETAILS

Removal of soot and noxious gases (NOx, HC, CO) from various potential sources (such as diesel engines) still represents one of the major task for a significant improvement of the quality of air. It is particularly interesting and is the aim of this project to propose the fundamental study of mechanistic aspects involved in the processes of catalytic combustion of soot in the presence of oxygen and/or of other pollutants such as NOx, CO, etc. This requires an in-depth knowledge of the chemistry and behavior of the oxidation process of the particulate matter, by identifying the key parameters that rule the process as well as a detailed knowledge of the catalytic materials to be proposed and exploited. One of the final targets is the development of new catalysts able to take advantage of the oxidant ability of the NOx, present in the exhausts gases, for the soot combustion.

Traditional LNT (Lean NOx trap) catalyst for the simultaneous removal of NOx and soot (named DPNR) have been already proposed by Toyota and have formulations like $PtBa(K)/Al_2O_3$ (LNT catalyst). Such a new catalytic converter for DPNR is a porous ceramic wall-flow filter coated with a LNT catalytic layer. These catalytic systems work under cyclic conditions: during lean phase the NOx produced by the engine are oxidized and subsequently adsorbed on the alkaline or earth-alkaline metal oxide component (with nitrite-nitrate species formation); during rich phase the nitrate-nitrite species are reduced to molecular nitrogen by CO, H_2 and unburnt HC. On the other hand, the particulate matter removal takes place under both lean and rich conditions. The reactivity of a typical $Pt\text{-}Ba(K)/Al_2O_3$ LNT catalyst in the simultaneous NOx and soot removal shows good performances towards the removal of both pollutants. When alkaline and noble metals are present, a synergistic effect has been pointed out in presence of NO; accordingly, a specific role of superficial species (e.g. nitrates or other surface species formed in the presence of NOx) having high activity in the soot oxidation can be invoked. Actually a complex interaction between catalyst, particulate, gas-phase NOx and stored NOx has been indicated as responsible for synergistic effects which promote the combustion activity at low temperatures.

One of most important drawbacks of LNT systems is their deactivation caused by sulphur poisoning due to the formation of sulphates on the catalyst surface that reduced its storage capacity. Recent studies have proposed ceria-based materials as innovative LNT catalytic materials for the NOx removal. These systems showed high de-NOx activity even at low temperatures (200°C) and also good resistance to sulphur deactivation. Indeed the use of supports based on cerium oxide confers interesting properties to soot combustion catalysts due to high availability of surface oxygen and high surface reducibility. Accordingly, these systems, eventually doped with noble metals and alkaline and/or alkaline-earth metals, would represent an effective option as innovative DPNR catalytic systems able to reduce NOx and soot emissions. To our best knowledge, it has never been reported in the literature an investigation on the combined soot-NOx removal activity according to the DPNR concept over ceria based catalysts which is one of the targets of this proposal.

Object of this research project will also be the study of combined LNT-SCR systems, which have been recently proposed in order to overcome the problem related to ammonia slip observed in the LNT systems. It has been recently recognized that ammonia formation over the LNT catalyst during the rich phase can be exploited as NOx reductant when an ammonia-SCR catalyst (active in a low temperature region) is coupled with the LNT catalyst. In fact ammonia which is released from the LNT system during rich conditions can be stored on the SCR catalyst which is located downstream the LNT layer. Significant increases in efficiency of NOx removal and selectivity are reported for such combined systems. The depth knowledge of the LNT systems will therefore rationalize the operation of such combined systems: in fact, despite the advantages of the combined use of LNT + SCR systems are well recognized, still many aspects of the reaction mechanisms that operate during both lean

and rich conditions of these configurations require further clarification. Finally, the activity of such combined systems in the simultaneous removal of NOx and soot will be studied.

The research activity will be organized in different phases:

- Selection of early catalyst formulations.
- Reactivity and fundamental study of NOx sorption on the selected catalysts.
- Development of a reaction cell hosting catalytic filters
- LNT+SCR combined system investigation: activity tests and kinetic studies.