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# Ceria-praseodymia nanostructured catalysts for NO, soot and NOx-assisted soot oxidation: from powder to wall-flow reactor

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***Highlights***

- Equimolar nanostructured ceria-praseodymia has greater soot oxidation activity than ceria
- Ceria-praseodymia outperforms Pt on ceria in NOx-assisted soot oxidation
- Ceria-preaseodymia confirmed its reactivity also when deposited in a wall-flow filter

## 1. Introduction

The work includes the preparation of nanostructured ceria-praseodymia impregnated with small Pt nanoparticles stabilized by n-octylsilane. 1.0 wt.% loading of Pt nanoparticles on ceria-based materials have been achieved by wet impregnation on pure ceria (Ce-NP) and ceria-praseodymia (Ce50Pr50-NP) nanoparticles (hereafter called Pt/Ce-NP and Pt/Ce50Pr50-NC). The idea behind these multicomponent systems stems from the fact that both the Pt and the Ce-Pr equimolar mixture are effective towards NO and soot oxidations, as well as for the $NO_x$-assisted soot oxidation. The equimolar Ce-Pr catalyst was finally deposited on a SiC wall-flow filter, for soot cake regeneration.

## 2. Methods

The synthesis involves a ripening – precipitation process [1], starting by dissolving equimolar Ce and Pr nitrate precursors in 8M NaOH solution, carried out in an autoclave kept at 180 °C for 24 h. The Pt nanoparticles are separately obtained as a dark brown colloid by mixing Pt(dba)2 and Si(n-C8H17) in dry THF under Ar, which is then exposed to 3 bar of $H_2$ [2]. 1%-wt loading of Pt was achieved by wetness impregnation (WI) of the catalytic supports. Finally, the Ce50Pr50-NP were deposited by a dipping process, through repeated immersions of the bare SiC filter in the catalyst-containing slurry, and calcinations at 650°C, able to reach a catalyst loading of 15%wt in the filter. A soot loading of 6 g/l (i.e 1%wt on the filter) and a GHSV=15000 $h^{-1}$ were used in the structured reactor oxidative tests.

## 3. Results and discussion

Ce-NP has a low surface area of 8 $m^2$ $g^{-1}$from BET characterization, while Ce50Pr50-NP has higher surface area, namely 16 $m^2$ $g^{-1}$. Figure 1 shows TEM images of Pt/Ce-NP (Fig.1A-1C) and Pt/Ce50Pr50-NP catalysts (Fig.1D-1F). The deposition of small Pt nanoparticles (2 nm) went onto Ce-NP surface, while the deposition on Ce50Pr50-NP results in a less homogeneous particle distribution. Figure 2A shows the CO conversion over the prepared catalysts during the test with CO oxidation (an intermediate of soot oxidation). Ce50Pr50-NP exhibits higher activity (lower temperature range) than pure ceria. The activity of Pt-containing catalysts is evidently higher than that of Pt-free catalysts. Figure 2B shows the $NO_2$ percentage as a function of temperature during the test with NO oxidation. Pure ceria Ce-NP sample has evidently the lowest catalytic activity as the reaction lights off at ca. 350°C and reaches its maximum conversion at 500 °C. Pt/Ce-NP gives a very early reaction light-off and maximum conversion at 400 °C.

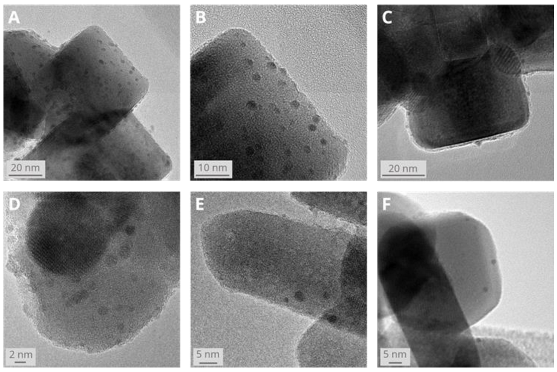

**Figure 1**. TEM images for the prepared samples.

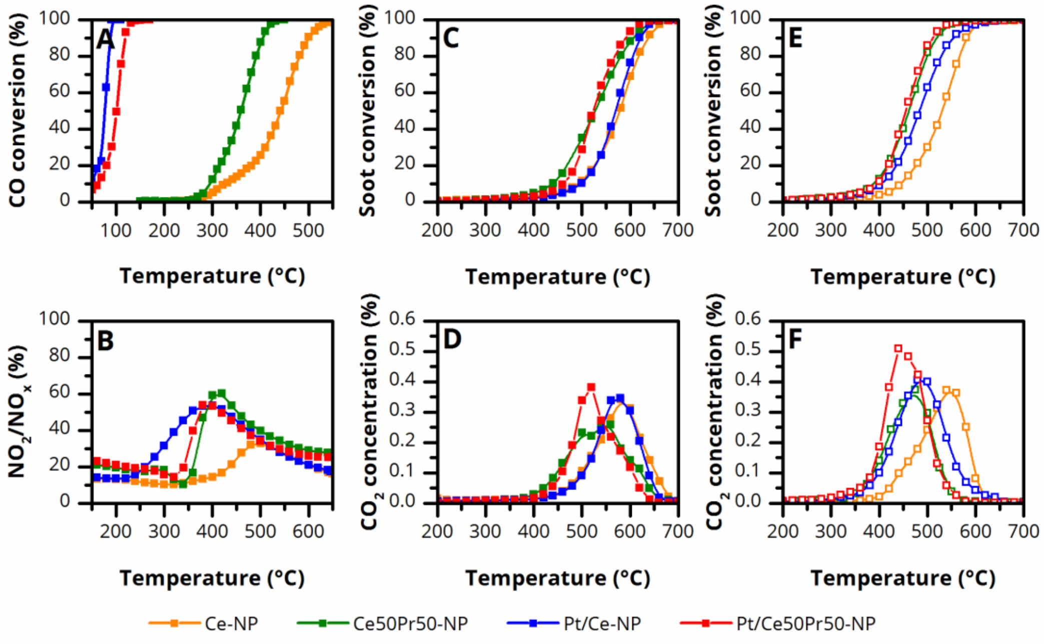

**Figure 2**. Performances of the catalysts during the tests for the (A) CO oxidation, (B) NO oxidation as $NO_2$ percentage, (C – D) soot oxidation and (E – F) $NO_x$-assisted soot oxidation, in terms of total soot conversion (C and E) and $CO_2$ emission (D and F).

Ce-Pr samples present a unique profile as they accommodate $NO_2$ adsorption at low temperature (300–350°C) –the results are obtained in a temperature raping test, and were also verified at steady state isothermal experiments–. Maximum NO conversion can be very high in Ce-Pr systems; Ce50Pr50-NP in particular has higher NO conversion than Pt/Ce-NP. The addition of Pt on the surface of Ce50Pr50-NP appears to give small benefits in earlier reaction light-off. Figure 2C and 2E shows the soot conversion during the test with *$NO_x$-free* and *$NO_x$-assisted soot oxidation*, respectively: Ce50Pr50-NP (green curve) outperforms Pt/Ce-NP (blue curve). This might be because Pr not only facilitates NO oxidation to $NO_2$ but also enables strong $NO_2$ adsorption. The adsorbed $NO_x$ species, fixated on the surface, can be in direct contact with soot in soot-catalyst interphase, thus initiating the oxidation process.

The Ce50Pr50-NP catalyst was finally deposited on the filter wall surfaces, and tested against soot oxidation: in brief, in a filter regeneration operated isothermally at 400°C for 4h, the $NO_x$-free soot conversion was 83%, against a value of 76% obtained with pure Ceria. The $NO_x$-assisted soot oxidation with Ce50Pr50-NP increased soot conversion up to 88%, thus confirming its superior performance also in structured form.

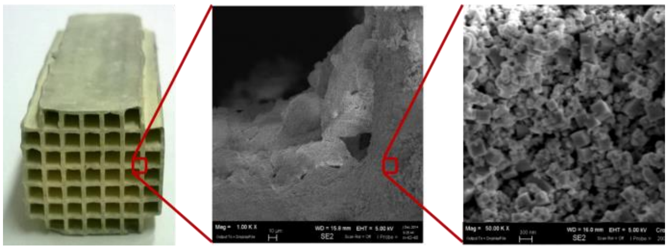

**Figure 3**. Ce50Pr50-NP monolith synthesis.

## 4. Conclusions

Catalytic activity tests have shown the effectiveness of nanostructured Ce50Pr50-NP towards the $NO_x$-assisted soot oxidation. From powder and reactor scale tests, the addition of Pt on the Ce50Pr50-NP surface appears unnecessary, except for $CO_2$ selectivity improvement.

## References

[1] Andana, Piumetti, Bensaid, et al. Applied Catalysis B 197 (2016) 125-137.
[2] Andana, Piumetti Bensaid, et al., Applied Catalysis B 209 (2017) 295-310.

***Keywords***
Soot oxidation, NOx, nanostructures, filter regeneration, Ceria, Praseodymia.