Supported gold perovskite catalysts for diesel soot oxidation and CO emission control during DPF regeneration

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Introduction

An intensive research has been carried out in the last decade to find catalysts active for the abatement of diesel exhaust pollutants. The main contaminants emitted by this type of engine are nitrogen oxides and soot particles. The current tendency is to minimize NO_x production by high exhaust gas recirculation (EGR) rates and tackle the problem of high diesel particulate emissions with the combined use of traps and oxidation catalysts [1]. Diesel particulate filters (DPFs) based on wall-flow-type monoliths are generally recognized as the most viable solution to the related pollution problem [2]. Anyway the uncontrolled catalytic regeneration of a DPF showed high emissions of CO and $PM_{0.1}$ [3]. Since supported gold catalysts and perovskite catalysts show high catalytic activity towards CO oxidation [4] and soot combustion [5], respectively, the development of a multifunctional Au-perovskite catalyst for a controlled DPF regeneration was investigated with this work.

Materials and Methods

A series of perovskite catalysts ($LaMnO_3$, $LaCrO_3$, $LaFeO_3$, $LaNiO_3$) was prepared via a highly exothermic and self-sustaining reaction, the Solution Combustion Synthesis (SCS) method [6]. The Au-perovskite catalysts (Au-$LaMnO_3$, Au-$LaCrO_3$, Au-$LaFeO_3$, Au-$LaNiO_3$) were prepared via the same route, by adding $HAuCl_4$ to the precursors solution in order to obtain a catalyst with 2 wt% of gold. Characterization by FESEM-EDS, XRD and BET techniques was accomplished. The activity of the prepared catalysts was analysed by TPC, according to standard operating procedures reported in [5] for soot combustion and in [6] for CO oxidation, respectively. The $LaNiO_3$-2%Au catalyst was then deposited by *in situ* SCS directly over a SiC wall-flow filter and tested over real diesel exhaust gases with the procedure reported in [3].

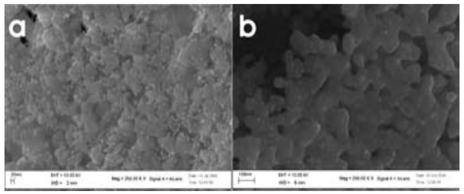

Figure 1. FESEM picture of: a) the $LaNiO_3$-2%Au catalyst; b) the $LaNiO_3$-2%Au catalyst deposited on the SiC wall-flow filter.

Results and Discussion

The SCS method has been developed to produce in a low-cost "one-step" process, homogeneous, very fine, crystalline powders without the intermediate steps which other conventional synthesis routes would require. Such features were also interesting for the co-synthesis of the supported Au catalysts; actually it was possible to achieve in one-shot Au clusters of comparable size (5-10 nm) with those obtained by more complex controlled precipitation methods [4]. Figure 1a illustrates a FESEM picture of the $LaNiO_3$-2%Au catalyst. It regards the catalyst which showed the highest activity among those prepared. Some data regarding the catalyst characterization are listed in Table 1 (CO_2 peak temperature T_p for soot combustion, and half- or full-conversion ones for CO oxidation). By comparing the activity of the considered catalysts towards CO oxidation it is clear that the presence of gold strongly enhances the rate of CO oxidation in all cases. Conversely, as expected, this positive synergetic effect cannot be observed for the activity towards the carbon combustion which is governed by perovskite-carbon direct contact points. Figure 1b shows a FESEM picture of the $LaNiO_3$-2%Au catalyst deposited on the SiC wall-flow filter. The catalyst on the filter maintains the same nanostructure observed for powder catalysts. Figure 2 compares the results of the loading and regeneration runs [3] for the $LaNiO_3$-2%Au-catalyzed and the virgin wall-flow traps. The results are rather encouraging. The regeneration of the catalyzed trap was much faster than that of the bare ceramic filter. Significant amounts of fuel can thus be saved during filter regeneration, thereby reducing the operating costs of the soot abatement system. Moreover a significant difference was noticed when analyzing the CO concentration during the regeneration process: the $LaNiO_3$-2%Au catalyst enabled a CO abatement higher than 85 %. A "mild" regeneration run was also carried out with low fuel post-injection showing negligible formation of secondary $PM_{0.1}$ nanoparticles [3].

Table 1. Collection of results of catalyst characterization tests concerning catalytic activity and BET surface area.

Catalyst	BET [m^2/g]	C oxid. T_p [°C]	CO oxid. T_{50} [°C]	CO oxid. T_{100} [°C]
$LaMnO_3$	16.4	524	254	315
$LaMnO_3$-2%Au	17.7	525	234	290
$LaCrO_3$	15.8	506	450	580
$LaCrO_3$-2%Au	22.2	505	285	500
$LaFeO_3$	29.4	524	290	340
$LaFeO_3$-2%Au	18.7	525	215	240
$LaNiO_3$	14.5	430	242	280
$LaNiO_3$-2%Au	10.4	431	156	197

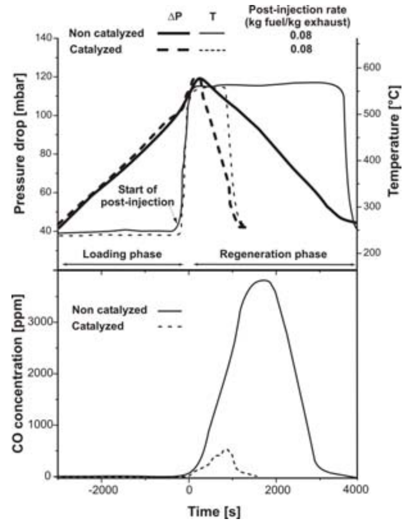

Figure 2. Loading and regeneration runs for the $LaNiO_3$-2%Au-catalyzed and the virgin wall-flow traps.

References

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