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Catalytic performances of ceria and ceria-zirconia materials for the combustion of diesel soot under NO_x/O_2 and O_2. Importance of the cerium precursor salt

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ABSTRACT

Two different cerium precursors ($Ce(NO_3)_3{\cdot}6H_2O$ or $(NH_4)_2Ce(NO_3)_6$) and two different calcination temperatures (500 °C or 1000 °C) were used in the synthesis by coprecipitation method of $Ce_xZr_{1-x}O_2$ mixed oxides with three different Ce/Zr ratios ($Ce_{0.8}Zr_{0.2}O_2$, $Ce_{0.5}Zr_{0.5}O_2$ and $Ce_{0.2}Zr_{0.8}O_2$) along with pure cerium and zirconium oxides, obtaining a wide range of oxides with very different textural and structural features. These catalysts were tested for soot combustion in loose-contact mode both under NO_x/O_2 and O_2 atmosphere, pointing out that the choice of the $(NH_4)_2Ce(NO_3)_6$ precursor leads to mixed oxides with better catalytic activity in the two atmospheres proved with regard to the $Ce(NO_3)_3{\cdot}6H_2O$ precursor. Catalysts with ceria-rich compositions showed better catalytic behaviour than the corresponding zirconia-rich compositions, demonstrating that BET surface area is not the only parameter to be considered for this application, since the catalytic activity of the prepared samples is not proportional to surface area of the solids. Important contribution of the NO_2-assisted mechanism exists both to initiate and continue soot combustion under loose-contact mode in NO_x/O_2, when the ceria-based material produces NO_2 at low temperatures (425–450 °C), being the capability of the catalyst to produce NO_2 the most important feature to explain the soot combustion. However, this role becomes minor when the ceria-based material produces NO_2 at higher temperatures. Under O_2, in a loose-contact situation, a good Zr-insertion into the lattice affects positively to the soot combustion activity.

1. Introduction

The different legislations dedicated to diesel particulate matter (denoted as PM) emissions of vehicles (i.e. for Europe, 0.005 g of particulates/km for passenger cars since 2009) have led to the development of different post-treatment technologies. These are based on the use of a filter (the most common type is a wall-flow filter of either silicon carbide or cordierite) [1,2]. Once the particles are collected, it is necessary to burn them off. Diesel particles spontaneously burn in air at about 500–600 °C. This temperature range is not regularly achieved in the typical diesel vehicle applications for sufficient periods of time to enable self-regeneration [1]. Therefore, a controlled regeneration must be conducted in order to avoid back pressure and sudden burn-off, which might occasionally cause temperature to raise above the melting point of the filter [3].

Considering practical conditions, two main options are proposed for the regeneration of the filters: (i) periodic increase of temperature by heat injection [4–6] (i.e. with an electrical heater or by fuel injection) or (ii) decrease of the soot combustion temperature with a solid catalyst. Concerning the second approach, there are two different alternatives: the so-called fuel borne catalysts (addition of a catalyst precursor, usually a soluble organometallic compound, to the fuel [7,8] and loading a catalyst coating onto the filter [1,9]. EURO V regulations are forcing to adopt the latter solution. Presently, most car manufacturers admit the necessity of a combined use of catalysts and particulate traps [1].

Ceria-based oxides and, in particular, ceria-zirconia mixed oxides are among the most appropriate materials to fulfil the strict requirements that a soot combustion catalyst should possess: acceptable activity in the exhaust temperature (usually below 500 °C), good thermal shock resistance, stability in the presence of steam, improved thermal stability against sintering compared to that of pure ceria and so on [10–25].

There are many parameters that can strongly influence the ceria materials' performance as soot combustion catalyst. Under loose-contact of catalyst and soot, the ceria-soot interface will play a major role since the contact is very poor [17,26–29]. In this case, a successful catalyst design will demand a deep knowledge of the relationship existing between Ce-Zr composition/atomic homogeneity/textural features (active surface area exposed)/structure (crystalline phase/s) and the soot combustion activity [27,30].

Most of our previous studies concerning catalysed soot combustion were carried out under NO_x/O_2, since NO_2 is determinant for initiating and continuing soot oxidation in the loose-contact mode,

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