



Constructing nano-structure on silver/ceria-zirconia towards highly active and stable catalyst for soot oxidation

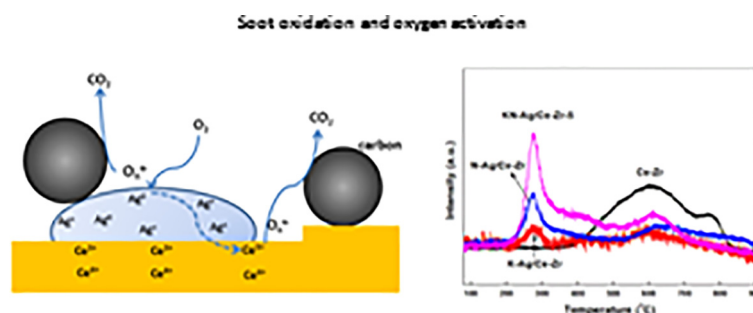
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HIGHLIGHTS

- Highly active Ag/Ce-Zr catalyst with 'Ag/Ce' structure was successfully synthesized.
- Precipitant can affect the size of Ag particles and alter Ag-Ce interaction.
- The optimal interaction was beneficial for the increase of superficial Ag⁺ species.
- The catalyst/soot contact for the Ag/Ce-Zr catalyst is expected to be enhanced.
- Ag/Ce-Zr catalyst was utilized five times without any appreciable loss of activity.

GRAPHICAL ABSTRACT



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ABSTRACT

Different to Pt-based catalysts used in diesel particulate filter (DPF), Ag/CeO₂-ZrO₂ catalyst, whose main feature is a considerable amount of silver species exposed on the surface of catalyst, possesses outstanding soot oxidation performance even in loose contact mode and in the absence of NO_x. The Ag/CeO₂-ZrO₂ catalyst with the special structure and Ag particle ranging from 17 to 35 nm is herein prepared by controlling precipitant, and its combustion temperature T_{10} and T_{max} can be achieved as low as 254 °C and 280 °C in 5.0% H₂O/4.8% O₂/90.2% N₂ and loose contact mode, respectively. The H₂-TPR and XPS characterization results show that the interaction between Ag and ceria, controlled by precipitant, leads to the high content of the species of Ag⁺ and Ce³⁺. It can be concluded that the distribution of silver species with high superficial Ag⁺/Ag⁰ ratio and Ag-Ce interface in Ag/CeO₂-ZrO₂ catalyst does favor the activation and transfer of oxygen species. It appears that the adsorbed oxygen species play an important role in catalytic performance. Moreover, the catalyst/soot contact for the Ag/CeO₂-ZrO₂ catalyst is expected to be enhanced due to the transfer of oxygen species. The prepared Ag/CeO₂-ZrO₂ catalyst is repeatedly used five times without any appreciable loss of activity, thus shows good stability initially.

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1. Introduction

Soot particles emitted from vehicles especially diesel engines are one of the main sources of urban atmospheric particulate

matters, which cause acute human health and environmental problem. Currently, the most commonly used method for removing soot from engine exhaust is the use of diesel particulate filter (DPF), which collects soot followed by oxidation of the

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accumulated particulates at elevated temperature. The DPF usually works in an atmosphere containing a fairly low amount of oxygen and almost no NO_x , which can also be removed as advanced technologies developed [1–4]. Therefore, traditional Pt-based catalyst will not be effective under this circumstance.

Moreover, even in the case of using high cost platinum group metals as catalyst, the contact between the catalyst and soot in the catalyzed DPF is relatively loose, while the catalyst may not be well constructed to effectively transfer/diffuse activated oxygen species to reaction sites, resulting in low catalytic activity for soot oxidation on whole, i.e. requiring higher temperature to trigger the soot combustion. Such a temperature is generally measured and denoted as T_{10} defined as the temperature when 10% of total feeding soot is oxidized in test experiments. If the T_{10} takes more time to accomplish especially starting a cool engine, undesired soot accumulation happens, leading to increase the exhausting pressure and therefore decrease the engine efficiency. Once the accumulated soot is not effectively removed (oxidized) and high exhausting pressure appears, a compulsory de-soot procedure via burning intently injected fuel has to be carried out. Accordingly, there is a need to develop stable catalysts used in a DPF which contain little or no platinum group metals but highly active to effectively catalytic oxidized soot at temperature as low as possible. On the whole, a commercially viable soot oxidation catalyst should have high catalytic activity thus low soot ignition temperature and thermal stability as basic properties as the exhausting temperature will generally be raised as the engine works for some time.

The catalytic reaction of soot oxidation is a typical heterogeneous catalytic reaction, which takes place at the three-phase boundary among a solid catalyst, a solid reactant (soot), and the gaseous reactants (O_2 , NO_x etc.). The essence of soot oxidation is how to improve the catalytic oxidation activity for concerned catalysts, and construct more favorable structure for transferring/diffusing activated oxygen species to reaction zones on catalyst, and also promote better contact between catalyst and soot. The study of soot oxidation (combustion) is not only far-reaching for environment protection as applied in vehicle exhausts, but also fundamentally important in heterogeneous catalysis [5–8] as a solid-solid-gas multi-phase reaction.

First, it is fundamental to improve the oxidation performance of catalyst along with reversible pseudo-phase change of the active species [9–14], leading to decrease the soot ignition temperature generally called T_{10} defined as the temperature when 10% of total feeding soot is oxidized in test experiments and similarly for T_{50} and T_{90} corresponding to 50% and 90% soot conversions respectively. However, the species with excellent oxidation activity for soot oxidation catalysts are normally unstable thus lack of commercialization prosperity. A typical example is that Ag_2O , as the most active specie for soot oxidation among 30 tested metal oxides, showed high activity even under inert atmosphere, but completely lost the activity after one catalytic run [15]. For Ag-based catalysts, it is generally made use of interaction between silver and support to deal with this issue in previous works [16–21]. It was reported that Ag-based catalysts such as Ag/SiO_2 , Ag/CeO_2 , Ag/SnO_2 , $\text{Ag}/\text{Al}_2\text{O}_3$ possess relatively high oxidation activity of soot when Ag interacts optimally with its supports. It is suggested that ceria is shown to make silver stable in the oxide state, while with ZrO_2 and Al_2O_3 metallic Ag is preferentially formed [22]. In a word, the preparation method plays a vital role in their interaction thus the stability of catalyst. Moreover, there are quite a few studies focused on ceria-supported silver as soot combustion catalysts, since the ceria possesses excellent storage and transfer capacity for oxygen species [23,24].

Furthermore, it is proposed that an appropriate interaction or/and interfacial effect from Ag– CeO_2 catalysts can efficiently promote soot oxidation [15,20,21]. The interaction of Ag– CeO_2 catalyst

prepared by impregnation can improve its oxidation activity to some extent. However, this catalyst exhibited poor stability. Kayama et al. reported that aggregates of ceria particles around silver metal, i.e. Ag– CeO_2 with ‘rice-ball’ morphology, synthesized by a one-pot selective redox reaction, exhibited exceptional catalytic performance for soot oxidation by O_2 below 300 °C [20,21]. Thus, it is also generally considered as one of the most efficient catalysts for soot oxidation in absence of NO_x [2]. And the exceptional catalytic performance can be ascribed to the interfacial effect of Ag with ceria and the promotion of formation and migration rates of oxygen species on this catalyst. Moreover, as stated by Kayama et al., the special structure with character of ‘rice-ball’ morphology is suitable as a catalyst, in contrast to core-shell materials wherein the shell deactivates the catalyst metal. However, the related study on optimization of interfacial effect has not been subsequently conducted.

In addition, Ag– CeO_2 core-shell nano-spheres, whose particle sizes range from 30 to 100 nm, were fabricated by a surfactant-free method involved with a subsequent annealing redox reaction [25]. The as-prepared catalyst exhibited enhanced catalytic performance for CO oxidation due to strengthening of the interfacial interaction. One can see that the interfacial interaction of Ag with ceria has been investigated to a certain extent, most of the studies focused on the process of post-treatment, which could be too tedious and difficult to control in large scale preparations.

In a word, the Ag-based catalyst, being characteristic of exposing most of silver species on its surface, is seldom studied although it is paid more attention to the Ag– CeO_2 catalysts with ‘rice-ball’ or core-shell structure. Moreover, it is imperative to develop a facile method to accomplish the optimization of interaction between Ag and ceria from which the catalytic active species can be stabilized.

The improvement of contact performance is doubtlessly important in soot oxidation [26–31]. It is evident that the capacity of oxidation cannot be fully exploited, even the catalyst endowed with outstanding oxidation activity towards other reactions when the contact of soot/catalyst is not so appropriate. The problem is still a bottleneck for improvement of contact efficiency, thus in turn catalytic activity. To enhance the contact efficiency between soot and catalysts, molten salts catalysts or fuel-borne catalyst were used to wet the soot surface and therefore decrease the oxidation temperature [32,33]. It is expected that Ag– CeO_2 catalyst with optimized interaction may also enhance its contact with soot, and thus improve its oxidation activity.

Furthermore, although CeO_2 is characteristic of good oxygen-storage capacity, its tendency to deactivating at high temperature (through sintering and structure collapsed) impedes its practical application. Doping with other elements (ions) such as Zr^{4+} is an effective way to stabilize CeO_2 against sintering and collapsing [34,35]. The Zr-doped CeO_2 material not only possesses increased thermal stability, but also exhibits improved oxygen storage capacity. It is thought that this beneficial effect on the creation of oxygen vacancies is due to the partial substitution of Ce^{4+} ions with Zr^{4+} ions. Hence, CeO_2 – ZrO_2 was chosen as the support of Ag-based catalyst as it can at least provide a excellent thermal stable support for the catalyst.

Although several studies have been focused on the morphology of Ag– CeO_2 with core-shell or ‘rice-ball’ structure, the relatively fine structure of Ag– CeO_2 , beneficial for improving the efficiency of contact between catalyst and soot, has not been reported. In this study, the distribution of Ag species in particle of catalyst can be controlled just using different precipitant such as KOH or/and $\text{NH}_3 \cdot \text{H}_2\text{O}$. And even in the absence of NO_x , the as-prepared Ag-based catalysts exhibited excellent and considerably stable soot oxidation activity. Then, it was investigated the effect of precipitant on the distribution of silver, the reduction behavior of catalyst, the

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