

Ag-loaded cerium-zirconium solid solution oxide nano-fibrous webs and their catalytic activity for soot and CO oxidation



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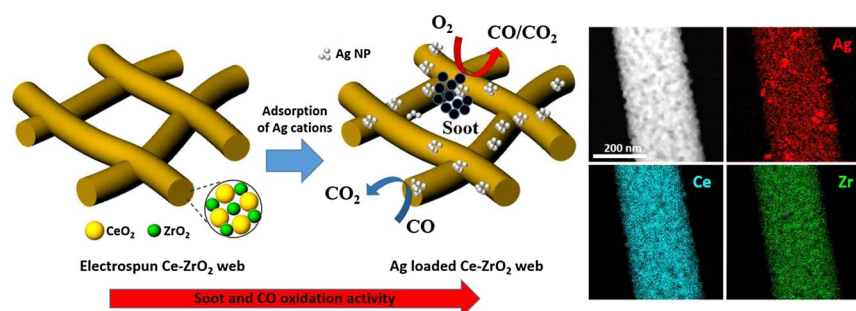
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GRAPHICAL ABSTRACT



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ABSTRACT

The catalytic combustion of soot and CO is one of the key technologies required to meet rigorous emission standards. Recently, solid solution materials have been employed in heterogeneous catalysts because of their remarkable intrinsic activities and good stabilities. However, the low number of contact points between soot particles and the catalyst remains a challenge to enhancing catalytic performance. Thus, we herein report the preparation of Ce-ZrO₂ solid solution nano-fibrous web catalysts with a hierarchical structure using an electrospinning method, where Ag particles were loaded onto the surface of the Ce-ZrO₂ webs. X-ray diffraction, scanning transmission electron microscopy, and energy dispersive spectroscopic studies allowed us to investigate the morphological and crystal structures of the prepared Ce-ZrO₂ and Ag/Ce-ZrO₂ web catalysts. Moreover, the relationship between the Ce/Zr ratio and activated oxygen is discussed based on X-ray photoelectron spectroscopy results. Following the catalytic oxidation of soot and CO using our novel materials, we found that the Ce_{0.67}Zr_{0.33}O₂ web exhibited higher catalytic activities than the Ce_{0.5}Zr_{0.5}O₂ and Ce_{0.33}Zr_{0.67}O₂ webs, respectively. In addition, Ag/Ce_{0.67}Zr_{0.33}O₂ exhibited enhanced catalytic activity compared with the pristine Ce_{0.67}Zr_{0.33}O₂ for the oxidation of both soot (e.g., 500 °C vs. 544 °C at 50% conversion) and CO (e.g., 282 °C vs.

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408 °C at 50% conversion). It therefore appeared that our proposed Ce-ZrO₂ solid solution nano-fibrous web catalysts bearing Ag particles exhibited superior redox properties and enhanced surface areas, and as such, are promising candidates for use in the oxidation of both soot and CO.

Nomenclature

TPB	triple phase boundary
3DW	three-dimensional web
PM	particulate matter
HAADF-STEM	high-angle annular dark-field scanning transmission electron microscopy
FE-SEM	field emission scanning electron microscopy
HR-TEM	high-resolution transmission electron microscopy
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction
TPO	temperature programmed oxidation

TPR	H ₂ -temperature-programmed reduction
TCD	thermal conductivity detector
TGA	thermogravimetric analysis
CB	carbon black
LC	loose contact
T _i	starting temperature of soot ignition
T ₅₀	50% point of soot conversion temperature
T _f	finished temperature of soot oxidation
SAED	selected area electron diffraction
BET	Brunauer-Emmett-Teller
BJH	Barrett-Joyner-Halenda

1. Introduction

Diesel engines have attracted significant attention for use in automotive vehicles due to their enhanced fuel efficiency compared to gasoline engines. However, the particulate matter (containing mostly soot particles), carbon monoxide (CO), nitrogen oxide (NO_x) and unburned hydrocarbons expelled from the fuel combustion process can cause respiratory diseases in addition to environmental pollution [1]. Currently, a continuously regenerating particulate trap has been suggested as an efficient aftertreatment technique that functions via a physical trap and a catalytic oxidation process [2–6]. Hence, numerous diesel oxidation catalysts have been intensively studied, and these are generally composed of precious metals supported on oxide materials [4]. Indeed, in recent years, precious metals (e.g., Ag, Au, Ag-Au alloy, and Pd) have been widely used to superior catalytic performances and stabilities for the oxidation of soot and CO [7–11]. In addition, Ag-modified catalysts have recently been suggested for application in catalytic reactions because of their relatively low costs and significant performance improvements, which are related to the generation of superoxide species

[12,13]. As the catalytic properties of support materials are crucial for determining both catalyst stability and catalytic activity, various support materials, such as alkali-metal oxides [14–16], spinel oxides [17,18], perovskite-related oxides [19–21], and ceria-based materials [22–24], have been developed. Of these, ceria-zirconia (Ce-Zr) solid solutions are frequently considered as promising support candidates for soot and CO oxidation catalysts because of their excellent redox properties and high oxygen storage capacities, in addition to their excellent thermal stabilities [25–27]. However, as few studies have been carried out into Ag-loaded Ce-Zr supports, it is particularly important to gain an understanding of the interactions taking place between Ag and the Ce-Zr support to achieve enhanced catalytic activities for the oxidation of soot and CO.

The activity of a heterogeneous catalyst is mainly influenced by the number of contact points between the solid reactant (soot particle) and the catalyst surface. Moreover, the small pore sizes (< 10 nm) of these catalysts presents a challenge for achieving high catalytic activities, as soot particles (> 25 nm) cannot infiltrate the inner pores of the catalysts [28]. To address these issues, novel strategies that can increase the

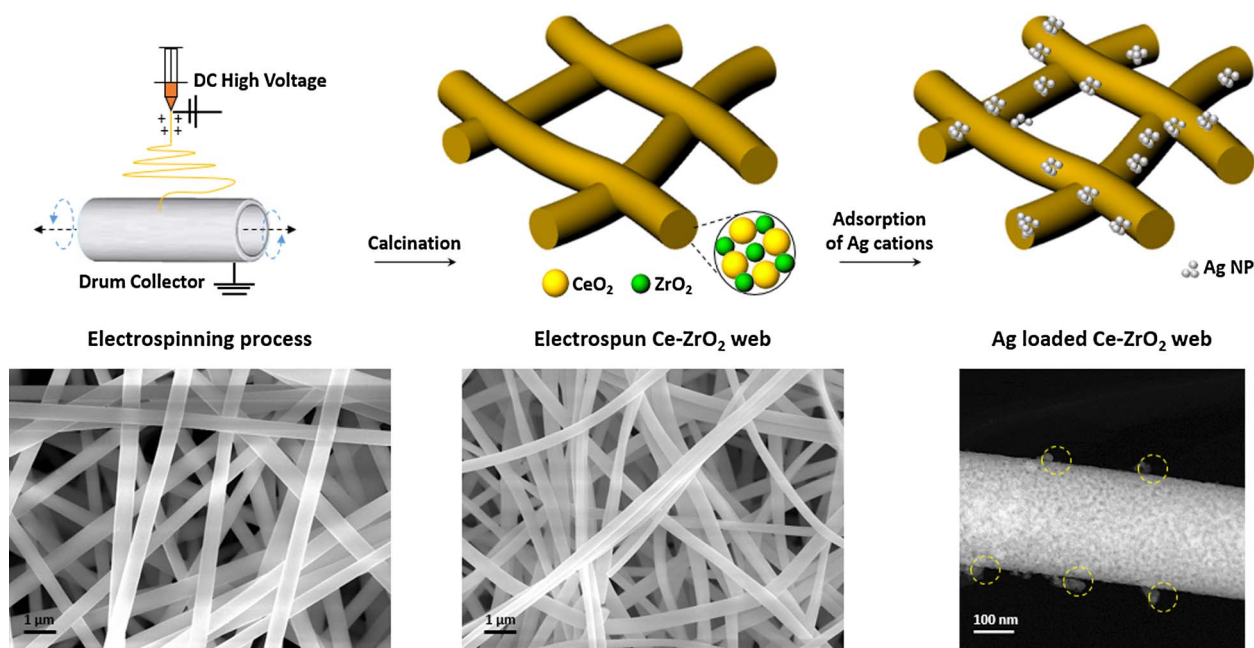


Fig. 1. Overall procedure to prepare Ag/Ce-Zr fibrous webs via typical electrospinning and deposition processes.

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