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Effect of lanthanum loading on nanosized CeO₂-ZnO solid catalysts supported on cordierite for diesel soot oxidation

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ABSTRACT

We report the application of a solid lanthanum–ceria–zinc catalyst in the catalytic regeneration of diesel particulate filters (DPF) in diesel engines. We synthesized a CeO₂-ZnO-La₂O₃ (Ce–Zn–La) mixed oxide by a lactic acid-mediated sol–gel method, which efficiently coated cordierite substrates for soot capture and combustion. We studied the effects of La loading on the physicochemical and catalytic properties of Ce–Zn mixed oxide during low-temperature soot combustion processes. We characterized the synthesized catalysts by X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), N₂ adsorption, Raman spectroscopy, oxygen storage capacity (OSC), and scanning and transmission electron microscopy (SEM and TEM). Thermogravimetric and differential thermal analysis (TGA/DTA) confirmed that the catalysts effectively reduced the soot oxidation temperature. The ternary Ce–Zn–La mixed oxide catalyst with Ce/Zn/La atomic ratio of 2:1:0.5 had the highest catalytic activity and promoted soot oxidation at temperatures below 390°C. This indicated that the large number of oxygen vacancies in the catalyst structure generated oxygen species at low temperatures. Raman spectroscopy measurements revealed the presence of oxygen vacancies and lattice defects in Ce–Zn–La samples, which were key parameters concerning the stability and redox properties of the prepared catalysts.

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Introduction

Particulate matter (PM) is one of major pollutants in diesel exhaust and is abundantly present in urban environments. Removing diesel exhaust products is essential to mitigate the health and environmental issues associated with PM, mainly in large cities (Zhang et al., 2015; Godoi et al., 2016). PM consists primarily of soot covered with adsorbed aromatic

hydrocarbons, and it causes cardiovascular and respiratory diseases in urban populations (Wang et al., 2017; Reşitoğlu et al., 2015; Moldanová et al., 2009; Arteaga et al., 2015; Wen et al., 2017). Post-treatment systems can generally remove such harmful substances. Improvements in these systems have helped to meet the increasingly stricter environmental regulations regarding diesel and pollutant gas emissions. The main strategy to retain soot particles from diesel exhaust

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gases involves monolithic wall-flow type filters, which typically consist of open monoliths where gas mixtures are forced to flow through porous walls in small parallel channels that are alternately plugged at their ends (Nascimento et al., 2016).

Cordierite honeycombs ($2\text{MgO}\cdot 2\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) are excellent ceramic substrates to produce diesel particulate filters (DPFs): they have high melting point, great thermal shock resistance, excellent chemical stability, and good catalyst adhesion (Yang et al., 2017; Soloviev et al., 2015). Depending on the size and amount of channels and on gas percolation through the walls, ceramic honeycomb filters provide high filtration efficiency. However, the filtration mechanism causes soot to deposit when the filter is saturated, which can lead the vehicle combustion system to fail, lose power, and consume more fuel (Nascimento et al., 2016). This happens because the particulate material ignition temperature commonly lies between 600 and 700°C, whereas the exhaust temperature of diesel engines typically reaches less than 500°C under normal working conditions. Thus, filters need to be periodically regenerated in oxidizing, high-temperature conditions or continuously regenerated in the presence of suitable catalysts that can oxidize soot at exhaust working temperatures (Tang et al., 2014; Nascimento et al., 2014a; Kumar et al., 2012).

CeO_2 -based materials display very high catalytic activity towards diesel soot oxidation (Trovarelli and Fornasiero, 2013). Nevertheless, their poor thermal stability results in sintering and decreases the surface area, which is a serious drawback (Trovarelli and Fornasiero, 2013; Atribak et al., 2008). To overcome this problem, ceria has been partially substituted with a wide range of metal cations of different oxidation states. Modifying ceria to obtain mixed oxides stabilizes the catalyst against sintering and enhances the redox efficiency. This is possible because oxygen vacancies are created or oxide ion mobility increases, to culminate in higher oxygen storage capacity (OSC) (Bueno-López, 2014; Aneggi et al., 2014; Nascimento and Serra, 2016).

Mixed metal oxides are very interesting materials for catalytic oxidation/reduction reactions (Wang et al., 2016; Gao et al., 2017), water gas shift reactions (Si and Flytzani-Stephanopoulos, 2008), and solid oxide fuel cells (Ferreira et al., 2010). The precursors are inexpensive; their synthesis and substitution processes are simple and allow one to adjust the structure–property relationship to fit the target applications of the resulting mixed oxides (Wang et al., 2012). We have recently proposed that ZnO can act as a diffusion barrier to suppress CeO_2 sintering, to improve durability at high temperatures (Nascimento et al., 2014a). Other studies have shown that mixed oxides obtained from $\text{Ce}^{3+}/\text{Ce}^{4+}$ and Zr^{4+} precursors by the sol–gel methodology exhibit superior OSC (Nascimento et al., 2014b). In addition, a suitable technique to improve the activity of ceria-based catalysts is to create multifunctional catalysts containing inter-, multi-metallic or alloy compounds on the catalyst surface.

Interaction between oxide surfaces and catalytic metals typically increases porosity, which avoids segregation and creates more oxygen vacancies and defects in the catalyst lattice (Bueno-López, 2014; Aneggi et al., 2014). These metal compounds must be directly formed during catalytic system activation. These newly generated defects are expected to induce more surface-active oxygen species and to accelerate

oxygen exchange with the environment by a vacancy diffusion mechanism, to promote soot oxidation at lower temperatures and effective filter regeneration. These compounds are thought to be a key factor for the achievement of highly active redox reaction systems.

Recent literature attests that CeO_2 modification with rare earths and aliovalent transition metals increases the physicochemical stability and the OSC of the final catalyst (Gálvez et al., 2013). Accordingly, in the present study we have modified CeO_2 with both transition (Zn^{2+}) and rare earth (La^{3+}) metal ions by a sol–gel method, which was followed by annealing at 650°C. Then, we evaluated how the catalytic efficiency in soot oxidation depended on lanthanum addition to Ce–Zn mixed oxide. After deposition of $\text{CeO}_2/\text{ZnO}/\text{La}_2\text{O}_3$ ternary mixed oxide over cordierite, we conducted catalytic activities to investigate controlled surface and structural properties, to find out that the proposed system is a promising catalyst material for diesel soot particulate oxidation.

1. Experimental

1.1. Preparation of the sol gel mixed ternary oxide powder

The CeO_2 -ZnO- La_2O_3 ternary oxide (Ce–Zn–La) catalysts were synthesized according to the conventional lactic acid sol–gel method. An ethanolic solution containing a mixture of cerium(III) nitrate ($\text{Ce}(\text{NO}_3)_3\cdot 6\text{H}_2\text{O}$, 99%, Fluka), lanthanum(III) chloride ($\text{LaCl}_3\cdot x\text{H}_2\text{O}$, 99%, Merck), and zinc acetate ($\text{Zn}(\text{CH}_3\text{COO})_2$, 99%, Aldrich) was employed; the Ce/Zn/La molar ratio in the solution was 2:1:0.5, and the final metal concentration was 0.5 mol/L. Lactic acid (85% Acros) was used as a complexing agent. The mixture was heated under reflux. Lactic acid was successively added until the suspension was totally dissolved, giving rise to a stable sol (Nascimento et al., 2014a). Ethanol was partially eliminated in a rotary evaporator, and the gel product was dried at 70°C overnight. The sample was heated at 650°C for 4 hr under air atmosphere to obtain the solid catalysts.

1.2. Preparation of the Ce–Zn–La catalyst coated onto cordierite

The cordierite honeycomb monoliths (Umicore®, specific surface area < 3 m²/g) were used as DPF (cylindrical shape, 100 cells per square inch, diameter of 2.5 cm, and length of 3.5 cm). The monoliths were coated with the Ce–Zn–La-based sol by successive dipping at 40°C for 5 min. After each step, the coated cordierite was dried at 70°C for 4 hr and annealed at 650°C for 4 hr. This procedure was repeated five times to create an irregular surface with higher specific surface area. The amount of catalyst was 5%–6% in mass in relation to the initial mass of the cordierite substrate (Nascimento et al., 2014a, 2014b, 2016).

1.3. Characterization of the prepared materials

Specific surface areas (SSA) were measured from N_2 absorption-desorption isotherms acquired at liquid nitrogen temperature (Nova 2200, Quantachrome, USA) surface analyzer; the BET (Brunner–Emmet–Teller) method was applied. Before the measurements, the samples were heated under low pressure

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