



Development of a stacked wire-mesh structure for diesel soot combustion



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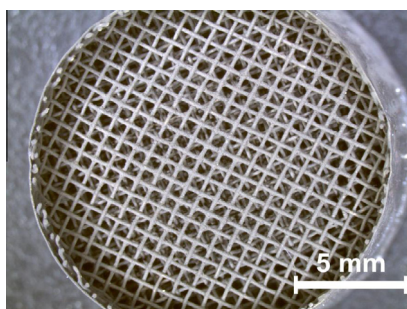
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HIGHLIGHTS

- New catalytic wire-mesh filters for diesel soot combustion.
- Metallic filters designed and built using wire-mesh discs of AISI 304.
- Excellent Co, Ba, K/CeO₂ catalyst washcoating.
- Wire-mesh geometry influences directly the coating adhesion of the catalyst.
- Very high activity at temperatures relevant for diesel soot abatement.

GRAPHICAL ABSTRACT

Catalytic diesel soot filter made of wascoated wire meshes.



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ABSTRACT

This work describes the development of stacked wire-mesh structures and their washcoating method to prepare a structured system for the catalytic combustion of soot. For this purpose, a metallic structured substrate was designed and built using wire-mesh discs of AISI 304. The coating of this substrate was optimized by investigating various strategies and parameters of the washcoating such as the suspension formulation (sol-gel coating with posterior impregnation of the active phase, the all-in-one strategy using catalyst precursors and the preparation of the catalyst slurry with a ready-made catalyst), the amount of Co, Ba, K/CeO₂ catalyst and the packing material (wire diameter and number of packed discs). The coating proposed strategies were analyzed measuring textural properties, catalyst coating adherence and morphology, and pressure drop of the catalytic substrate. Finally, the coated wire-mesh substrates were tested in the combustion of soot showing promising catalytic activity.

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1. Introduction

Diesel engines emit nitrogen oxides and soot that are produced during the fuel combustion [1]. Since both pollutants have negative environmental effects, research in this field has received increasing consideration in the last decades.

Many options for the catalytic removal of soot from diesel exhaust gases have been considered. The powder catalyst itself

cannot be used for a practical application in the diesel engine exhaust pipe due to the severe operating conditions of the high flows. Therefore, the catalytic filters coated with a catalytic layer are considered an attractive system. These structure substrates with filtration properties trap the soot particles and allow their burning at the exhaust gases temperature [2,3].

The most popular way to trap diesel particles is by means of surface filtration, sometimes known as a cake filtration or sieving [4], using monoliths made of cordierite [3–6] or silicon carbide [7,8]. These structures can be used as particulate filters when

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channels are alternatively plugged to force the diesel exhaust to flow through the porous walls of the monoliths and, in this way, soot particles are trapped. The wall-flow monolith filter acts as a cake filter, i.e. after some initial particle deposition into the pores of the monolith walls occurs, the cake layer formed becomes the dominant filtration medium [4,9]. But, in general, these monoliths offer significant pressure drop and narrow channel structures inside which a laminar flow takes place, and this fact implies mass-transfer limitation from the gas phase to the catalytic layer on the wall.

A second possibility is to use deep-bed filters using materials of relatively open structure compared to surface filters. In these filters, soot is retained throughout the filter structure by three well-known mechanisms: Inertial interception, Brownian diffusion and flow-line interception [4]. The main advantage of this strategy is the much better contact between the deposited soot and the catalyst coating of the filter, making it easier to regenerate the filter continuously. Deep-bed filters can be made of foams [9,10] or wire-meshes [11]. These substrates can be used in multifunctional reactors, which combine reaction and separation (filtration, adsorption), or combustion of feed mixtures containing gaseous and organic matter as particulates (soot) in a layered reactor [9,12,13]. During the last years foams and wire-meshes have not been considered as good filters [4]. However, Vanhaecke et al. [9] recently published that a β -SiC foam with pore diameter (window size) of $500 \pm 25 \mu\text{m}$ was effective in filtering ultrafine soot particles checked in a real bench test. Doggali et al. [13] also suggested that ceramic foams present especially potential option for old generation engines with very high PM emissions. Moreover, the efficiency of wire-mesh filters with window sizes similar to that of foams would be relatively low at the beginning, but during the particle deposition it would increase [14]. On the other hand, as Van Setten et al. [4] pointed out, the reported efficiencies are reasonable, especially when one considers that non optimized materials are used (standard foams are designed for other applications such as molten-metal filtration or structural material). Higher filtration efficiencies might be expected for tailor-made materials, among them wire-meshes, which can be found in many commercial sizes of wire and mesh openings.

In spite of the fact that there are few studies about wire-meshes for soot combustion, these filters present interesting properties to be used as substrates. The most attractive feature of the wire-mesh substrate is the price: the cost of these structured substrates is only about 25% that of ceramic honeycombs and foams [15]. There have been attempts to utilize wire-meshes made of cheap iron or stainless steel as supports for active catalyst component [16–18]. In addition, wire-mesh catalysts combine the excellent mass- and heat-transfer characteristics of pellets catalysts with a relatively low pressure-drop monolith, which is mainly attributed to the high porosity of the wire-mesh structures [12]. The radial mixing of the gases flow can readily occur through porous mesh structures yielding a more uniform distribution of fluids across the entire bed diameter [15]. Another advantage of the fibrous catalyst is its easy scale-up. The geometric flexibility of the wire-mesh catalyst makes it suitable for retrofit installations. It is thus possible to adapt the geometric appearance of the wire-mesh catalyst to almost any form required.

Numerous literature reports on the performance of wire-mesh catalysts study the influence of many design variables such as supporting grids, wire diameter, packing density, construction geometry (for honeycomb [17,18], rolling sheet [16] or packaged sheets [19]) and construction material (steel [16], nickel [20] or precious metals [21]). In order to prepare catalysts based on metallic wire-mesh, it is necessary to solve the problem of deposition of the catalytic layer (oxide support + active phase) over the metal wire. Since the thermal expansion coefficient of the metallic structured

substrate is different from the oxide support coating, the major problem is how to achieve a better adherence to the surface of the wire-mesh substrate. Various methods for obtaining well-adhered coatings on metallic substrates have been developed, including chemical vapor deposition (CVD), plasma spraying, washcoating and electrophoretic deposition [22,23]. But the preferred method for coating the oxide support powder is usually the conventional washcoating method, in which a fine thickness of catalyst support layer is formed on the surface of metallic substrate by repeatedly dipping the substrate into a slurry containing particles of the catalyst support materials, followed by drying and calcining. However, the authors that used washcoating methods for coating fibrous materials observed poor adherence and non-uniformity of the resulting coating [24,25]. Moreover, the coatings may flake away under the influence of mechanical stress [15]. On the other hand, few results have been reported about the coating adherence to wire-meshes using other coating methods. Ahlström-Silversand and Ingemar-Odenbrand [11] showed improved adherence of Al_2O_3 layers by the spray-deposition method, but other authors selected electrophoretic deposition of the Al_2O_3 layer as a pretreatment of the metallic wire mesh to generate a rough interlayer between wire-mesh surface and catalyst coating [17,20,26]. Nevertheless, this interlayer on wire-mesh is unnecessary when the catalyst is deposited by in situ growing [16,27] or using expensive metallic wire meshes that are active for the studied reaction [20,21].

For the ultimate goal of developing systems for the removal of diesel soot using wire-meshes, two major tasks must be addressed. One is the optimization of the filtering properties of the structure and the other, the effectiveness of the deposition of the catalyst active for the diesel soot oxidation on the wire meshes. This work focuses on the second one, where different strategies of coating are studied, along with the characterization of the produced structured catalysts and their catalytic properties for the oxidation of diesel soot.

2. Experimental

2.1. Construction of the wire-mesh structured substrate

Different wire-mesh discs of AISI 304 stainless steel (Filtración Vibración S.L.) were used in this work. The main characteristics of these wire-meshes are presented in Table 1. Fig. 1 shows the stages of construction of the structured substrate. The first stage was the building of a spot-welded cylindrical cartridge of AISI 304 stainless steel ($50 \mu\text{m}$ thickness, Goodfellow) of 16 mm in diameter and a height that depended on the number of wire-mesh discs of the structure (5–40 discs, which resulted in substrates 6–30 mm in height).

The wire-meshes were cut and corrugated (Fig. 1) in a home-made embossing tool (Fig. 1a), in this way allowing discs to remain slightly separated when stacked. Corrugated discs were 90° alternately stacked to prevent intermeshing within the metallic cartridge (Fig. 1d) and two mesh covers were also spot welded to lock the assembly (Fig. 1e). The substrate was then washed first in detergent–water and then in methanol, during 30 min in each solvent, in an ultrasonic bath.

Given its simplicity and versatility, the washcoating method was chosen to deposit the catalyst onto the structured supports. The pre-treatment of the metallic substrate before washcoating was necessary because the anchoring and interlocking of the washcoat particles with the surface irregularities produced in the pre-treated metallic substrate played an important role on the adherence of the catalyst to the structured substrate [28,29]. As reported, most of the coated metallic wire-meshes prepared without pre-treatment showed poor adherence [20,24,25]. Yang et al. [20]

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