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Study of the catalyst load for a microwave susceptible catalytic DPF

Vincenzo Palma [a,*], Paolo Ciambelli [a], Eugenio Meloni [a], Agusti Sin [b]

[a] Department of Industrial Engineering, University of Salerno, Via Ponte don Melillo, 84084 Fisciano (SA), Italy
[b] Pirelli Eco Technology, Via Luraghi snc, 20020 Arese (MI), Italy

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ABSTRACT

The development of a fast, safe and cost effective Diesel Particulate Filter (DPF) regeneration procedure is the major remaining technological challenge in the use of this device. In our previous works we showed that the simultaneous use of a MW applicator and a specifically catalyst loaded DPF, with 15%wt of $CuFe_2O_4$, allows to reduce the temperature, the energy and the time required for the DPF regeneration. Starting by these very promising results, in this work we continued to study in order to further improve the performances of the catalyzed DPF in terms of catalytic activity, to reduce the temperature and the MW energy required for the regeneration. The objectives of this work are to optimize the preparation procedure of the catalytic DPF, to study the effect of the active species load, and to verify the feasibility of the MW technology by assessing the energy balance of the regeneration phase, comparing it to the actually employed regeneration technologies. In the future we want to evaluate the activity toward the other pollutants present in the diesel exhausts (such as NO_x).

1. Introduction

Motivated by increasingly stringent emissions regulations, Diesel Particulate Filters (DPFs) have seen widespread use as the only technically and economically feasible means for meeting current and future Particulate Matter (PM) emissions limits [1].

Depending on the filtration mechanism, the DPF's may be classified as wall-flow and flow-through [2]. Flow-through type filters, made of ceramic foam, wire mesh or metal wool (Fig. 1), create a smaller pressure drop which in turn benefits the fuel efficiency but at the cost of a lower filtering efficiency, often below 60% [2]. The wall-flow type filters (Fig. 2) are honeycomb monoliths made of ceramic materials such as cordierite and silicon carbide which consist of a series of parallel channels alternatively plugged at each end to force the exhaust gas flow through the porous filter wall [3]: they are characterized by a filtration efficiency higher than 95% [4] and so they are the most effective system to control the soot emissions in Diesel engines [5].

During the filtration, the porosity is progressively blocked by the trapped soot particles, causing a growing pressure drop: so the collected soot inside the trap has to be periodically oxidized to regenerate the DPF. The oxidation step is promoted by the so-called 'passive' (at temperatures upstream the DPF of about 200–400 °C) and 'active' regeneration (temperatures upstream the DPF of about 550–600 °C) [6]. The exothermic PM combustion leads in some cases to excessive temperature rise, which may damage the ceramic DPF. The development of a cost effective, fast and safe regeneration procedure is the major remaining technological challenge in the use of this device [7]. Recently, there has been a lot of interest in using microwave energy to accelerate chemical reactions due to the rapid and selective heating of material through differential MW absorption [8].

These features are also considered to be particularly suitable for soot combustion in a SiC DPF due to the instantaneous penetration of microwaves into the filter body, the selective absorption of microwaves by the soot layer [9], and the good SiC dielectric properties, as evident from Table 1.

As indicated in Table 1, SiC seems to be the most adequate filter material. In addition, from the same data of Table 1 it is evident that the soot is also a very good MW absorber characterized by a high value of the Dielectric constant ε', and a very high Dielectric loss factor ε''. Furthermore, formulating the soot oxidation catalyst to absorb MW, the combination of MW heating with catalytic combustion may result in the effective oxidation of diesel soot at lower temperature and higher reaction rate [10–12].

The results of our previous deposition and on-line regeneration tests on uncatalyzed and Copper-Ferrite loaded DPF, showed that the simultaneous use of the MW and the catalyst loaded filter at lower gas flow rate, allows to reduce the energy supplied and the regeneration time than that required for the uncatalyzed filter [12]. In this work we propose to optimize the preparation procedure of the catalytic DPF in order to maximize the load of the Copper-ferrite on the filter in agreement with a sustainable pressure drop, and to verify the feasibility of this technology by assessing the energy

* Corresponding author. Tel.: +39 089 964147; fax: +39 089 964057.
E-mail address: vpalma@unisa.it (V. Palma).

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Fig. 1. "Flow trough" type filter.

Table 1
Dielectric properties of various materials [8].

Material	Dielectric constant ε′	Dielectric loss factor ε″
Diesel soot	10.70	3.600
Quartz	3.80	0.001
Cordierite	2.90	0.140
Alumina ceramic Al_2O_3	8.90	0.009
Silicon carbide SiC	30.00	11.000

balance of the regeneration phase, in order to compare it to the above described actually employed regeneration technologies.

2. Materials and methods

The DPFs used in our work are Pirelli Ecotechnology SiC wall-flow monolith filters with 150 cpsi, wrapped in an heat expanding intumescent ceramic-mat (Interam by 3 M) and enclosed in a stainless steel wave guide: in Table 2 are reported the physical and geometrical characteristics of the filters.

2.1. Catalyst preparation

As mentioned in the introduction paragraph, an active and stable soot oxidation catalyst if specifically formulated to absorb microwaves may combine microwave heating with catalytic combustion reaction for the effective abatement of diesel soot. It has

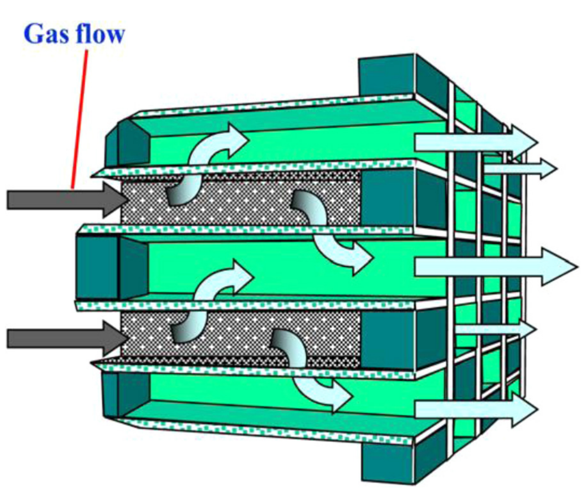

Fig. 2. "Wall Flow" Filter.

to be noted that due the microwave frequency values, on a macroscopic scale, a good number of catalysts can be considered a good microwave receptor, especially when they contains metal oxide having a large number of polar hydroxyl groups [10].

In the literature, the activity of catalysts based on copper and iron oxides toward soot oxidation in the presence of microwaves was studied: in particular Ma et al. [9] performed experiments of soot oxidation with and without catalyst using external heating to elucidate the influence of microwave irradiation on catalysis. They found that iron and copper were the catalysts most active in lowering the ignition temperature of diesel soot, while palladium was a necessary component in achieving a more complete combustion. In addition, the iron containing catalyst was very effective and energy-efficient at low microwave input.

In order to optimize the performance of microwave-assisted catalytic combustion of soot with a catalytic filter, which is related to a high capability in microwave dissipation, the complex permittivity of such a material must be taken into account, as deriving from the complex permittivity of the all filter components, i.e. soot, catalyst, support [10]. In previous works, the regeneration of a ceramic foam filter for soot trapping at the exhaust of a gas–oil burner had been performed in a specially designed single mode microwave cavity. The presence of catalyst was shown to enhance the soot oxidation rate in all the temperature ranges investigated [13,14].

The selected catalyst is based on the $CuFe_2O_4$, due to its very well known dielectric properties and good oxidation activity [12].

3. Results and discussion

3.1. Catalyst preparation

The Copper Ferrite ($CuFe_2O_4$) is prepared starting from iron nitrate ($Fe(NO_3)_3 \cdot 9H_2O$), copper nitrate ($Cu(NO_3)_2 \cdot 3H_2O$), mixed in a 2:1 molar ratio, and distilled water, continuously stirred at 60 °C. The catalytic DPFs have been prepared by repeated impregnation phases in the prepared solution, drying at 60 °C and calcination at 1000 °C after each impregnation, in order to obtain a 15%wt, 20%wt, 25%wt and 30%wt load of active species. Differently from the previous preparation procedure [12], we lowered the drying temperature to 60 °C and changed the calcination step (abrupt heating to 1000 °C, slow cooling to 500 °C and then quenching to room temperature): infact in the conditions previously developed and optimized for the lower catalyst load, we observed that at higher catalyst load the pores occlusion occurs, together with the occurrence of filter fractures which compromised its use, as also shown in literature [15]. With this new procedure, we realized a more uniform and homogeneous distribution of the active species on the DPF walls and inside the porosity (and not only on the channels external surface), reducing the occlusion of the inner walls pores, allowing to increase the catalyst load up to 30%wt.

3.2. Active species adherence testing

The adherence of the active species to the filter was evaluated measuring the weight loss caused by exposing the catalyst loaded monoliths to ultrasound, following the experimental procedure reported inn literature [16]. In this work the samples have been immersed in a beaker containing petroleum ether (Carlo Erba), and the whole was placed in an ultrasonic bath CP104 (EIA SpA) filled with distilled water, at a temperature of 25 °C, operating at 60% of rated power, for 30 min. Before the test, compressed air was blown through the monoliths in order to remove any possible residue. The weight changes were recorded during the test at regular intervals of 5 min after monoliths drying at 120 °C and cooling up to room temperature.