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Deep filtration and catalytic oxidation: an effective way for soot removal

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Abstract

Catalytic and uncatalytic axial flow ceramic foam traps for soot removal were prepared and tested at the exhaust of a gas oil burner. Soot filtration efficiencies of uncatalytic and catalytic traps were comparable and depended on the burner operating conditions. A threshold temperature of the catalyst of about 330 °C was determined. Above such a temperature while the pressure drop through the uncatalytic trap increases continuously because of soot load, that through the catalytic trap reaches a steady-state value where the soot amounts captured and burned on the trap were equivalent. Also radial flow ceramic foam traps were prepared and tested at the exhaust of a *common rail diesel* engine. Their behaviours reflected those of the corresponding axial flow traps. A model descriptive of the relevant phenomena (filtration, combustion and pressure drop time evolution) occurring all over the traps was formulated. It is able to yield the overall trap performances although further improvements are needed in the catalytic case. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Deep filtration; Catalytic oxidation; Diesel engine

1. Introduction

Diesel engine vehicles are widely used for their characteristics of economicity, simplicity of the engine itself, safety in case of accident. However, they have a major drawback since the soot particulate contained in the exhaust is claimed to be a potential carcinogen [1]. Both engine modifications and exhaust after treatment by filtration are worldwide utilised for the particulate matter reduction [2–7]. Engine modifications based on *common rail* technology, characterised by higher fuel injection pressure and better control of the injected fuel volume, greatly contributed to conform to the legislative demand. Nevertheless, the goal of matching the EURO IV limits of emissions (for vehicles of weight higher than 1600 kg) is strongly related to the availability of efficient catalytic de-NO_x and particulate filter systems [8,9]. To this purpose catalysts active for soot oxidation in the range of temperatures 300–400 °C have been proposed [3–6,10–13]. However, catalytic filters for diesel exhaust must fulfil other requirements crucial for the application such as: (i) good thermal shock resistance, (ii) low pressure drop, (iii) high soot filtration efficiency, (iv) high soot–catalyst contact efficiency [14]. Both wall flow and ceramic foam catalytic filters were investigated by us for particulate abatement at the exhaust of combustion systems [14,15]. The peculiar structure of ceramic foam filters, in particular, allows uniform particulate distribution along the device, resulting in homogeneous thermal load during trap regeneration, so limiting thermo-mechanical stress and risk of failure [16]. Furthermore, due to the deep filtration mechanism, the contact between soot and catalyst is favoured. Finally, since the cell size of ceramic foam is larger than pore size of ceramic honeycomb, a milder increase of

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engine backpressure is expected during soot trapping [17].

In this paper, the experimental results obtained treating the exhaust of a gas oil burner by axial flow ceramic foam uncatalytic and catalytic traps and the results of an overall model of traps performance are reported. In addition, preliminary results of performance tests of radial flow ceramic foam uncatalytic and catalytic traps at the exhaust of a new generation common rail diesel engine are also presented.

2. Experimental

2.1. Preparation of foam traps

Two types of traps were built to be tested at the exhaust of a gas oil burner and of a common rail diesel engine, respectively. In the former case axial flow traps while in the latter case radial flow traps were prepared. Therefore, 11 alumina foam disks (75 mm diameter, 7 mm thick) with 91% porosity and 65 ppi were assembled together to obtain the 0.34 dm^3 uncatalytic trap for burner exhaust treatment. Eight alumina foam disks (OD = 177.8 mm, ID = 30.48 mm, 15.24 mm thick) with 91% porosity and 65 ppi were put together to obtain the 2.94 dm^3 uncatalytic trap (Fig. 1) for diesel engine exhaust treatment. The catalytic traps of both types were made by assembling the disks as for the uncatalytic traps. However, in the former case the disks were previously loaded with Cu/V/K/Cl catalyst, through several cycles of impregnation with aqueous solution of catalyst precursor salts, drying at 120 °C and calcination at 700 °C overnight [12].

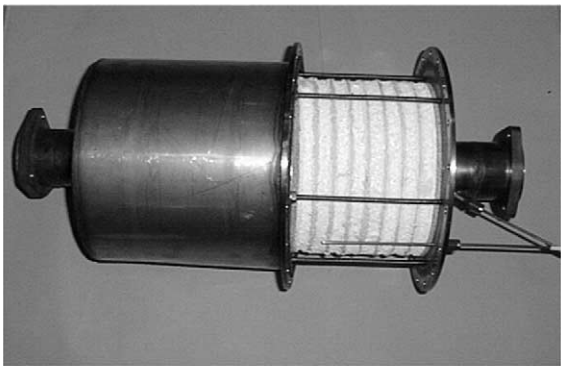

Fig. 1. Image of radial flow foam trap.

2.2. Apparatus set-up for burner exhaust treatment

The apparatus for trap testing includes a gas oil burner for soot generation fed with 1.9 kg/h commercial gas oil (H/C molar ratio of 1.75, S content of 0.05 wt.%). The air flow rate to the burner was varied in the range 28.5–45.6 m^3/h (STP) to obtain an air/fuel mass feed ratio (α) ranging from 23 to 37. The burner exhaust was fed to a stainless steel trap holder (20 cm long, 10 cm ID) located at 4 m far from the burner outlet. More experimental details are reported in [18].

2.3. Test procedures at the burner exhaust

Continuous filter operation was performed on catalytic and uncatalytic foam trap at different burner operating conditions (α between 23 and 37) and filter temperatures. The filter performances were evaluated by monitoring: (i) the pressure drop through the filter (ΔP), and (ii) the soot removal efficiency (η) by measurements of soot concentration upstream and downstream the trap with an Opacimeter (Tecnotest).

2.4. Apparatus set-up for diesel engine exhaust treatment

A 1.91 dm^3 JTD 16 valves four cylinders diesel engine, equipped with a common rail (Bosch) injection system with a maximum injection pressure of 1400 bar was employed. Trap temperature, filter pressure drop and soot removal efficiency (Opacimeter AVL 439) were measured.

2.5. Test procedure at the diesel engine exhaust

Three consecutive tests were performed with both catalytic and uncatalytic foam trap under the engine operating conditions reported in Table 1.

In order to prevent possible disk breaking, in these tests the ceramic foam disks were assembled together by gluing each other with ceramic cement so to obtain a rigid single monolith. In addition, the filter was enclosed in the stainless steel filter holder by interposition of a sealing vibration-damping gasket (Interam XD 3M).

Table 1
Operating conditions in the tests at the diesel engine exhaust

Test	Shaft speed (rpm)	Torque (N m)	P_{me}* (bar)	EGR duty (%)	Fuel mass rate (kg/h)	Air flow rate (STP) (m^3/h)	α	Soot concentration (mg/m^3)
1	1500	76	5	35	3.1	82	31	11.5
2	1500	136	9	0	5.2	93	21	55.4
3	2400	260	17	0	14.8	202	16	12.3

* P_{me}: mean effective pressure.

3. Model of filter performance

The overall model of filter performance comprises three sub-models taking into account: (a) the soot filtration; (b) the soot combustion over the filter surface; (c) the evolution of pressure drop through the filter as consequence of soot filtration and combustion, as well changes in gas mass flow rate or temperature.

(a) Soot particles in diesel exhaust can be treated as an aerosol. Relevant aspects of foam filters removal efficiency are: (i) capture of aerosol by the foam surface, acting as a collector; (ii) retention of captured aerosol on the surface collector and (iii) re-entrainment of retained soot as agglomerates, which can be brought away by gas. The overall net filtration efficiency is the product of the efficiency of particulate capture and the efficiency of retention of captured particulate on the collectors less the efficiency of loss of agglomerates in the leaving gas stream. Typical phenomena of conventional filtering, which lead to contacting and deposition of soot also occur on ceramic foam filters: diffusion, inertia (including gravity) and direct interception [19]. Filter performance is expressed in terms of the total capture efficiency of a single collector e defined as the ratio between the number of soot particles collected and the number of soot particles in the dusty gas approaching the collector. Efficiencies e_d, e_i, e_g, e_{di} are defined for each of the above filtering phenomena. Since for the filtration of aerosols by foams theoretical and experimental correlations for these efficiencies are not available in the literature, we have utilised correlations derived for fixed granular beds [20–22]. This was performed, however, by choosing the foam filter pore wall mean thickness as value of the key variable of these correlations, i.e. the mean particle diameter (d_p).

(b) This sub-model describes the interaction between catalytic surface and soot during the catalytic soot combustion process [23].

(c) This sub-model is based on the general equation for pressure drop through porous or granular systems, containing the viscous (linear) and the turbulent (quadratic) contribution term with respect to the superficial gas velocity (U):

$$\frac{\Delta P}{\Delta Z} = a_0^* U + a_1^* U^2 \qquad (1)$$

where ΔP is the pressure difference, ΔZ the filter thickness through which there is such a difference, and a_0 and a_1 are coefficients depending on the gas viscosity and density, on the bed voidage and on the specific filter surface area through two other coefficients β_1 and β_2, in turn depending on the system geometry. Values of specific surface area, of β_1 and β_2, suggested in the literature [24], were optimised for our specific filter.

4. Results and discussion

4.1. Performances of axial flow ceramic foam traps at the burner exhaust

Continuous tests lasting 2–3 h at the gas oil burner exhaust were performed by varying the values of either α and foam trap temperature. Typical results of tests with axial flow ceramic foam traps are shown in Fig. 2, where the percentage change of pressure drop through the uncatalytic and the catalytic foam traps $((\Delta P - \Delta P_0)/\Delta P_0)$, ΔP and ΔP_0 being the current and the initial filter pressure drop, are reported as a function of time at constant $\alpha = 37$. In the same figure the relevant filter temperatures are also reported. The pressure drop percentage deviation through the uncatalytic foam increases during the whole test but

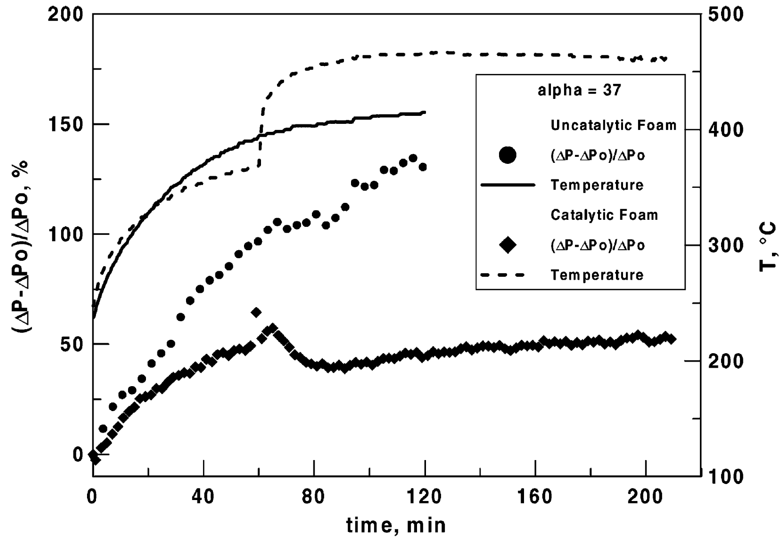

Fig. 2. Percentage deviation of pressure drop as function of time during continuous gas oil burner exhaust tests on catalytic and uncatalytic axial flow foam trap, $\alpha = 37$.

with a faster rate when the trap temperature increases and with a slower rate when the temperature reaches a constant value of about 400 °C. This gives evidences of the influence of temperature on pressure drop and of the continuous soot accumulation in the trap.

The pressure drop percentage deviation through the catalytic foam has a similar trend during the first 40 min when the temperature increases, but the rate of change is lower with respect to that of the uncatalytic foam. This difference indicates that in the former case the combustion of part of the filtered soot occurs at temperature higher than 330 °C. A further increase of temperature up to about 450 °C results firstly in a negative slope of $(\Delta P - \Delta P_0)/\Delta P_0$ then in a gradient almost equal to zero. This suggests that at these temperatures and $\alpha = 37$ the catalytic foam filter is active for soot combustion, being capable to burn an amount of soot greater than that captured and retained on it or, in other words, it is able to regenerate spontaneously.

The effect of the burner operating conditions on the trap performance is shown in Figs. 3 and 4, where the filtration efficiency and the corresponding temperature during tests at different α's with uncatalytic and catalytic trap are reported. Increasing α from 23 to 37, the filtration efficiency increases from 50 to 70% for both traps. This is likely due to the fact that increasing the excess of air results in a greater content of hydrocarbons adsorbed on the soot surface with consequent increase of soot adhesion to the filter surface. This leads to enhanced retention efficiency and, hence, increases the overall net filtration efficiency.

The comparison of results in Figs. 3 and 4 shows that the two traps exhibit different behaviour along the test duration: while the efficiency of the catalytic one is almost constant during the whole test, that pertaining to the uncatalytic trap increases as function of time. This increase is likely due to the decrease of foam porosity during soot accumulation, which determines an increase of both pressure drop and filtration efficiency. Instead, the lower amount of soot accumulated on the catalytic trap leads to a lower increase of pressure drop and an almost constant filtration efficiency.

4.2. Foam trap performance modelling

Fig. 5 reports a typical comparison of experimental and calculated time profiles of pressure drop through the uncatalytic foam. The agreement is good. In the same figure the linear and the quadratic contributions to the gas pressure drop (Eq. (1)) are also shown. They have similar values, as typical of low values of α. Increasing α and, hence, the gas flow rate the quadratic

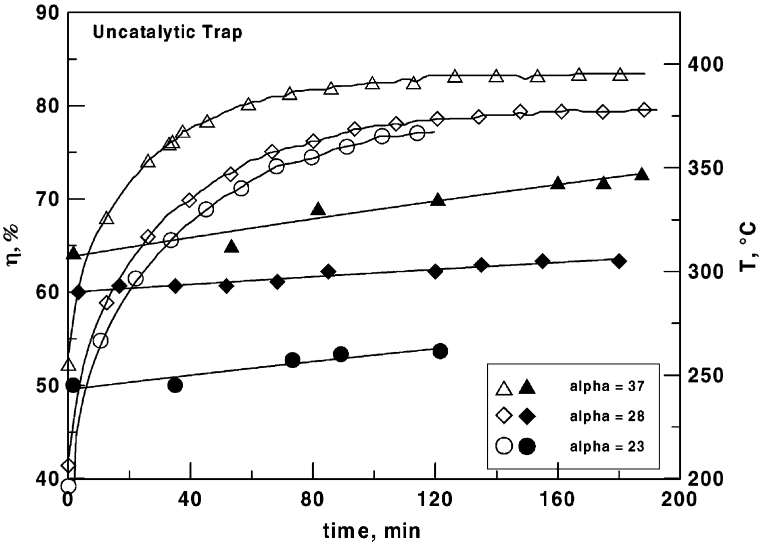

Fig. 3. Filtration efficiency and temperature of uncatalytic axial flow foam trap during tests at gas oil burner exhaust at various α. Full symbols: filtration efficiency; open symbols: temperature.

term becomes more and more prevailing, as expected. In any case, whatever α, the pressure drop profiles increase monotonically with time as the soot load over the filter increases. In Fig. 6 experimental and calculated time profiles of gas pressure drop through the catalytic foam relevant to α values of 23 and 37 are compared. With respect to those obtained with the uncatalytic foam, they reach an almost constant value after an initial increase associated with a temperature increase. This suggests that a balance between

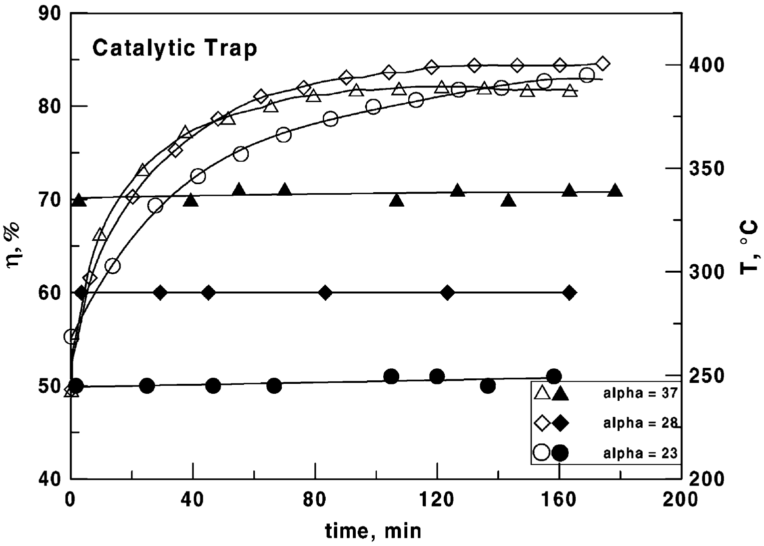

Fig. 4. Filtration efficiency and temperature of catalytic axial flow foam trap during tests at gas oil burner exhaust at various α. Full symbols: filtration efficiency; open symbols: temperature.

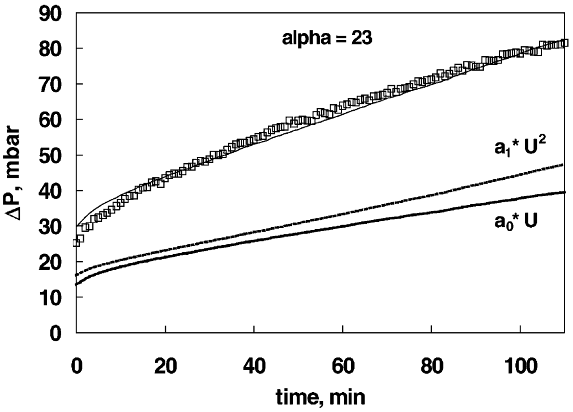

Fig. 5. Comparison between experimental (□) and calculated (line) pressure drop time profiles with indications, for this latter, of the linear and quadratic contributions (uncatalytic axial flow foam trap).

soot deposition rate and soot combustion rate establishes. Moreover, it can be noted that the lower α the lower the maximum of the gas pressure drop profile. This happens because at lower α the burner exhaust mass flow rate and, consequently, the mass flow rate through the filter is lower allowing a marked decrease of the gas pressure drop. In the specific case, changing α from 23 to 37, the exhaust gas mass flow rate increases from 45.9 to 72.2 kg/h. Comparison of Figs. 5 and 6 indicates that the agreement between model results and experiments is much better for the uncatalytic trap than for the catalytic one.

Model results also allowed the design of a trap optimised for lower pressure drop and with geometry suitable for vehicle applications. Therefore, a radial flow trap was realised for treating the diesel engine exhaust.

4.3. Performances of radial flow ceramic foam traps at the diesel engine exhaust

Preliminary results of tests with the uncatalytic and catalytic radial flow foam traps at the engine exhaust are shown, respectively, in Figs. 7 and 8, as time profiles of the gas pressure drop through the traps during the three consecutive tests. In the first test the results obtained in the catalytic and uncatalytic case are practically the same, the trap temperature being below the threshold catalyst temperature. In the second test the pressure drop of uncatalytic trap maintained slightly higher than that of the catalytic one. This difference can be attributed to the indirect effect of the catalyst on soot oxidation through the NO–NO_2 redox mechanism, as previously reported [25]. In the third test, instead, while the ΔP through the uncatalytic trap increases with time due to soot deposition (Fig. 7), the correspondent ΔP through the catalytic one first increases because of the sharp change of temperature, then decreases and, finally, reaches a constant value. It is worth noting that a ΔP decrease suggests

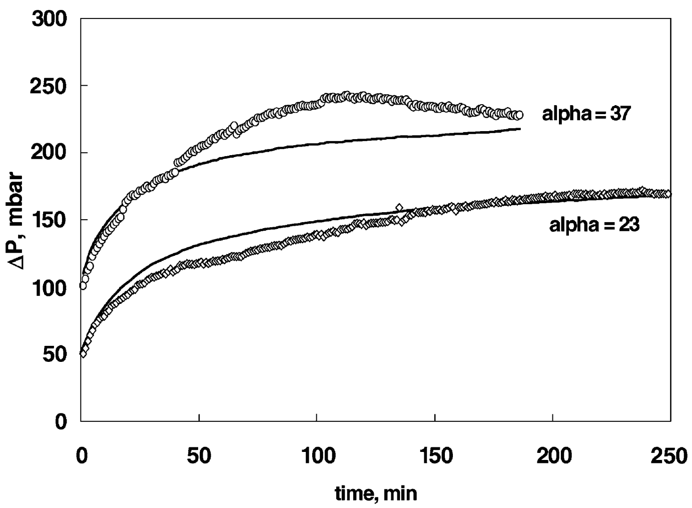

Fig. 6. Comparison between experimental (symbols) and calculated (lines) pressure drop time profiles for the catalytic axial flow foam trap.

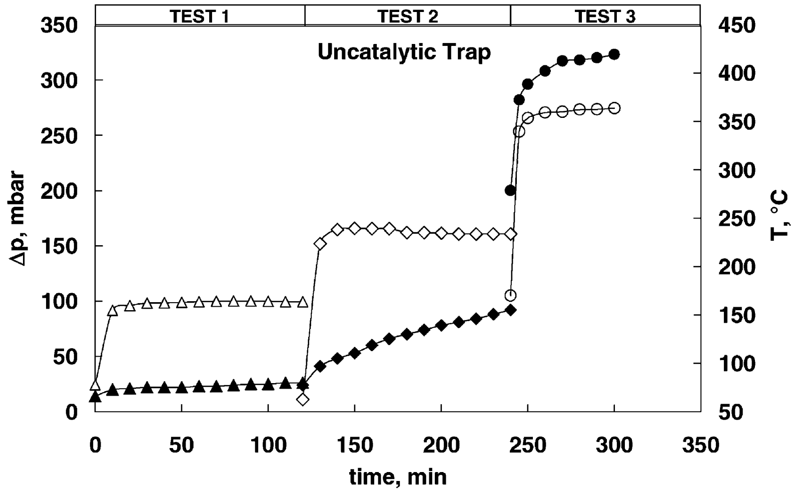

Fig. 7. Pressure drop and temperature of uncatalytic radial flow foam trap during tests at diesel engine exhaust. Full symbols: pressure drop; open symbols: temperature.

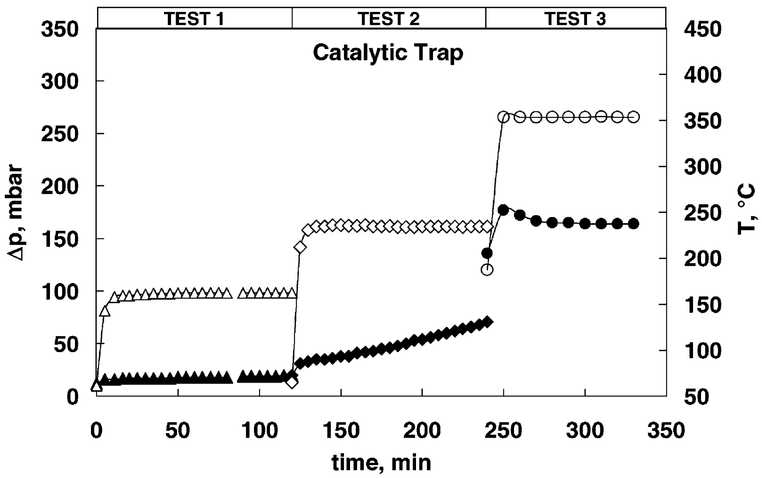

Fig. 8. Pressure drop and temperature of catalytic radial flow foam trap during tests at diesel engine exhaust. Full symbols: pressure drop; open symbols: temperature.

that the soot combustion rate is higher than the soot deposition rate over the trap and that a constant ΔP indicates that a balance between the soot deposition rate and the soot combustion rate is reached (Fig. 8).

Occasionally, during runs at higher flow rates with both catalytic and uncatalytic trap negative filtration efficiencies were found (test 3) likely due to the occurrence of soot blow-off phenomena.

5. Conclusions

Soot filtration and combustion are efficiently performed with alumina foam traps modified with Cu/V/K/Cl catalyst either at the exhaust of a gas oil burner or of a common rail diesel engine. Catalytic traps self-regeneration is achieved at 300–350 °C.

A descriptive model of the trap transient behaviour, helpful in optimising the trap design, was formulated. Fair agreement between model results and experimental findings were obtained although model improvements are needed in the case of the catalytic trap.

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