

Influence of Exhaust Gas Components on the Regeneration of Catalytic Ceramic Filters for Soot Removal

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The influence of exhaust gas components on the combustion of soot over unsulfated and sulfated catalytic filters was studied. The catalyst is deactivated by the presence of sulfur oxides in the exhaust gases, but it is able to regain most of its activity when it is employed in more realistic operating conditions. Indeed, with both unsulfated and sulfated catalytic filters, the presence of either NO or H₂O resulted in substantial increases in the rate of carbon combustion. The improvement in the soot catalytic oxidation rate in the presence of NO can be attributed to carbon oxidation by NO₂, which is, in turn, produced by NO oxidation on the catalyst. Instead, in the presence of H₂O, the observed enhancement could be due to the catalytic activation of the steam gasification of carbon on the same catalyst.

Introduction

Soot removal from Diesel exhausts can be accomplished with catalytic traps in order to match the increasingly severe limits for particulate emissions. In the past decade, the research efforts, devoted to the development of suitable oxidation catalysts with specific activity toward the total oxidation of the soot carbonaceous matrix, resulted in the formulation of very active catalysts in laboratory tests.^{1–13} In this context, a Cu/V/K/Cl catalyst supported on alumina powder (catalyst 137AA) was developed.^{3,6,9,14} In a few cases, catalytic filters were also tested directly at the exhaust.^{14–16} Moreover, only a few systematic investigations of the role or the effects of various exhaust gas components such as NO_x, SO_x, and H₂O were carried out,^{17–28} the main aim of those tests being the assessment of the system's overall performance.

It has been suggested that the soot combustion process either with^{17–24} or without^{25,26} catalyst can be improved by the simultaneous presence of O₂ and NO in the exhaust. It has been also reported that the simultaneous abatement of NO_x and soot could be achieved by a catalytic process in which soot is oxidized to CO_x and NO_x reduced to N₂.^{27,28} Recently, a systematic study of the effect of NO, SO₂, and H₂O on the catalytic combustion of carbon black²² on a Pt/SiO₂ catalyst showed that the presence of each of the three components above can increase the rate of carbon oxidation and that this effect is stronger when they are present together. However, the mechanism through which this occurs is not yet clarified.²² In this context, this work was focused on an investigation of the influence of the exhaust composition above on the regeneration of a catalytic filter prepared by catalyst deposition on commercially available ceramic filters. Regeneration tests of uncatalytic and catalytic filters, previously loaded with soot at the exhaust of a gas-oil

burner, were carried out in the presence of O₂; O₂ and NO; and O₂, H₂O, and NO in N₂ gas mixtures. The effect of filter sulfation on its regeneration was also investigated. Results were compared and discussed.

Experimental Section

Powder catalyst (named 137TF) was prepared by impregnating TiO₂ powder (DT51D, Rhône Poulenc) with an aqueous solution of NH₄VO₃, CuCl₂·2H₂O, and KCl (Baker Chemicals). The sample was then dried at 393 K and calcined at 973 K overnight.²³ A 2.9-cm-o.d., 1.3-cm-i.d. aluminosilicate tube (Goodfellow) with 50% porosity and 2.0 × 10⁻² cm mean pore size was divided into 3.0-cm-long sections. Catalytic (CF137TF) and uncatalytic filters were made by dipping the tube sections in slurries of 137TF/ethylene glycol and TiO₂/ethylene glycol, respectively. Filters were then dried at 393 K and calcined at 773 K for 2 h. Dipping in the TiO₂/ethylene glycol slurry was carried out prior to 137TF deposition so that the amount of catalyst deposited on the catalytic filter was about 3 g. The top side of each filter was then blinded with a 2.9-cm-diameter quartz disk glued to the filter by ceramic cement. A conical connection for the coupling with a specific settling in the quartz reactor was glued to the lower side of the filter as shown in Figure 1. The filter was then heated at 773 K for 1 h in order to allow the release of the organic components of the cement. Some samples of catalytic filter were severely sulfated before soot deposition and filter regeneration. The sulfation treatment, carried out at 693 K in a gas stream containing 1000 ppm SO₂, 10% H₂O, and 3% O₂ in N₂, took about 40 h. During this treatment, the filter adsorbed about 0.613 g of SO₂, corresponding to about 9 wt % of the total mass of TiO₂ powder and 137TF powder catalyst deposited on the filter. The thermal stability of the sulfur-containing species was checked by thermogravimetric (TG) analysis in N₂-O₂ flow in the temperature range 300–1000 K for a TiO₂/137TF powder catalyst mixture exposed to the same sulfation treatment as for the catalytic filter. TG analysis showed a 5.6% weight loss likely due to sulfite and/or sulfate decomposition in the

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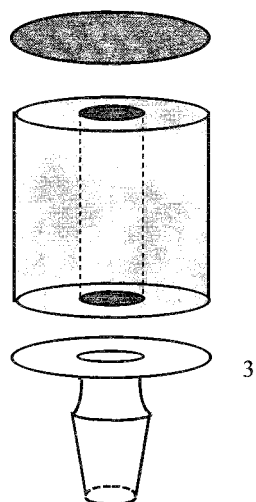


Figure 1. Sketch of the filter assembly: (1) quartz disk for top-side filter blinding, (2) filter, (3) conical connection for the coupling with a specific setting in the quartz reactor.

Table 1. Percentage Distribution of Main Catalyst Atomic Components Determined by EDAX Analysis of Powder Catalyst (137TF) (Fresh and Sulfated) and of Sulfated Catalytic Filter (CF137TF)

wt %	powder catalyst		sulfated
	fresh	sulfated	catalytic filter
Cu	8.1	10.6	10.1
V	7.9	6.2	6.8
K	7.0	8.0	5.9
Cl	5.2	0.0	0.0
S	0.8	8.9	5.8
Ti	71.0	66.3	71.4

temperature range 670–970 K. EDAX analysis, performed with a Philips XL30 scanning electron microscope equipped with an LaB₆ filament, showed the presence of Cu, V, K, and Cl in the fresh 137TF catalyst (Table 1). After sulfation of 137TF, EDAX analysis detected a complete substitution of chlorine by sulfur. Moreover, the relative amounts of the remaining significant elements were substantially unchanged except for a slight enrichment of Cu (Table 1). Table 1 also shows that sulfated 137TF powder catalyst and sulfated catalytic filter contain similar amounts of active elements.

Before each regeneration test, soot deposition over the uncatalytic or catalytic filter was carried out in an apparatus described in more detail elsewhere.^{23,24} It comprises a gas-oil burner for home heating acting as a soot generator, equipped with a nozzle giving a gas-oil mass flow rate of 0.53 g/s. Commercial gas-oil (H/C molar ratio of 1.75 and sulfur content of 0.05 wt %) was employed. The air flow rate to the burner was 9.5×10^3 cm³/s (NTP), and therefore, the air/fuel mass flow rate ratio (α) was 27.6. The soot-free filter was placed in a quartz tube (40-cm length, 3.5-cm i.d.), heated by an electrical furnace driven by a temperature-programmed controller (Ascon PY87). Then, a fraction [33.3 cm³/s (NTP)] of the burner exhaust stream, isokinetically sampled, was forced to pass radially through the filter to perform soot loading. The temperature of the sampling line was maintained at 423 K to avoid water and hydrocarbon condensation. During each deposition run, about 0.005 g of soot were collected on the filter.

After soot deposition over the filter, temperature-programmed oxidation (TPO) testing was performed with an apparatus described in detail elsewhere.⁹ The

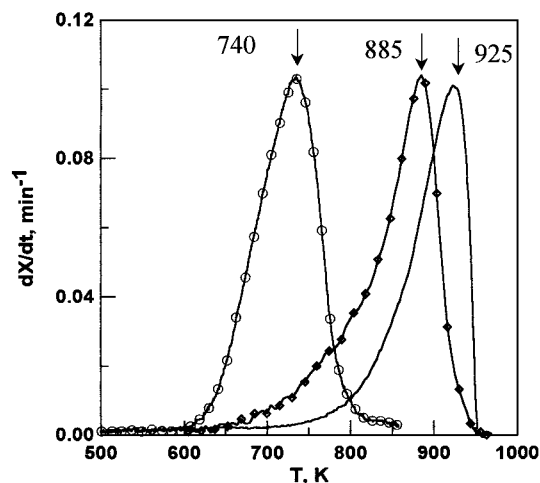


Figure 2. Reactivity–temperature profiles of catalyzed (CF137TF) and uncatalyzed soot combustion (feed gas = 10 vol % O₂, balance N₂; gas flow rate = 8.3 cm³/s (NTP); heating rate = 0.17 K/s). ○, catalytic filter; ◇, sulfated catalytic filter; —, uncatalytic filter.

filter was placed in a quartz tubular-flow microreactor (40-cm length, 3.5-cm i.d.) that was electrically heated to a controlled temperature (Ascon PY87). The reactor inlet gas flow rate was 8.3 cm³/s (NTP) of cylinder gas containing 10 vol % O₂ in N₂. The effect of the presence of secondary components in the feed gas was studied by performing TPO tests in which 1000 ppm NO, or 10 vol % H₂O, or 1000 ppm NO and 10 vol % H₂O was added to the reactor inlet gas stream. The temperature was increased at a rate of 0.17 K/s up to 873 K in the presence of catalyst and up to 973 K in the absence of catalyst while the operating pressure was 101 kPa in both cases. The tests were carried out with fresh or previously sulfated catalytic filters. NDIR continuous analyzers (Hartmann & Braun Uras 10E) measured the CO and CO₂ concentrations at the reactor outlet. Such concentration data were jointly integrated to calculate the overall conversion of carbon (\bar{X}) and the overall carbon reactivity ($d\bar{X}/dt$). \bar{X} was calculated as $(m_0 - m)/m_0$, where m and m_0 are the current and initial mass of carbon, respectively. Concentration data were also separately processed to evaluate the partial conversions to carbon monoxide (X_{CO}) and carbon dioxide (X_{CO_2}) and, therefore, the partial carbon reactivity (dX_{CO}/dt) and (dX_{CO_2}/dt). Carbon mass balance was verified within a 5% tolerance range for all tests. Two NDIR analyzers and a paramagnetic analyzer performed continuous monitoring of the NO, (NO + NO₂), and O₂ concentrations, respectively. All signals from the analyzers were acquired and processed by a personal computer.

Results

Figure 2 reports the comparison of the reactivity–temperature profiles relevant to TPO in an O₂–N₂ flow of soot deposited over uncatalytic and catalytic (fresh or sulfated) filters. It appears that the unsulfated catalytic filter markedly promotes soot combustion by lowering both the ignition and the burnout temperatures with respect to those for the uncatalyzed combustion. Correspondingly, the reactivity peak temperature was reduced by about 200 K. However, after sulfation, the catalytic filter activity dramatically decreased, giving rise to a 150 K backshift of the reactivity profile peak temperature ($d\bar{X}/dt$ curve), which approaches that of uncatalyzed soot combustion.

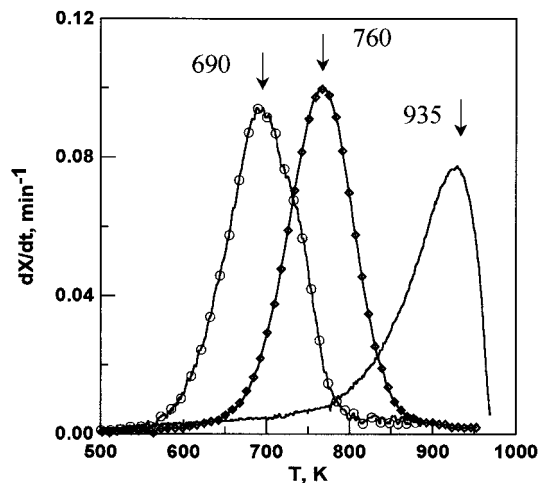


Figure 3. Reactivity-temperature profiles of catalyzed (CF137TF) and uncatalyzed soot combustion in the presence of H₂O (feed gas = 10 vol % O₂, 10 vol % H₂O, balance N₂; gas flow rate = 8.3 cm³/s (NTP); heating rate = 0.17 K/s). ○, catalytic filter; ◇, sulfated catalytic filter; □, uncatalyzed filter.

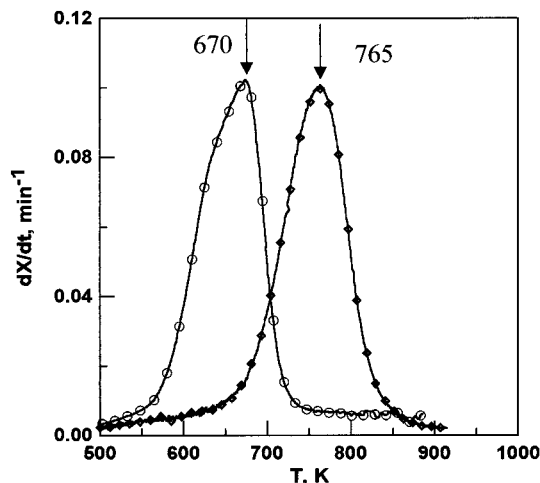


Figure 5. Reactivity-temperature profiles of soot combustion on unsulfated and sulfated catalytic filters (CF137TF) in the presence of NO + H₂O (feed gas = 10 vol % O₂, 1000 ppmv NO, 10 vol % H₂O, balance N₂; gas flow rate = 8.3 cm³/s (NTP); heating rate = 0.17 K/s). ○, catalytic filter; ◇, sulfated catalytic filter.

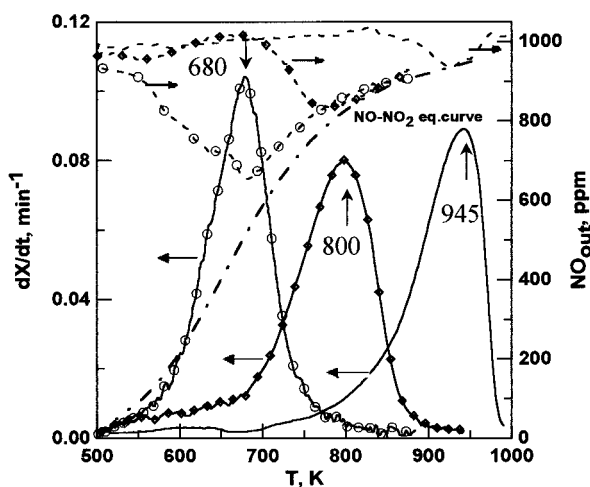


Figure 4. Reactivity-temperature profiles (continuous line) and relevant NO_{out} (dashed line) concentrations of catalyzed (CF137TF) and uncatalyzed soot combustion in the presence of NO (feed gas = 10 vol % O₂, 1000 ppmv NO, balance N₂; gas flow rate = 8.3 cm³/s (NTP); heating rate = 0.17 K/s). ○, catalytic filter; ◇, sulfated catalytic filter; □, uncatalyzed filter.

In Figure 3, the reactivity-temperature profiles relevant to soot TPO runs with catalytic and uncatalyzed filters in the presence of H₂O in the feed gas are shown. Whereas the presence of H₂O in the feed gas does not modify the reactivity profile of uncatalyzed soot oxidation, it strongly enhances the activity of the catalytic filter displayed with only O₂-N₂, especially in the case of the sulfated filter. Specifically, the soot reactivity peak temperature decreases by 40 K with the catalytic filter and by 113 K with the sulfated filter (see Figure 3).

Figure 4 shows the results of TPO tests of soot deposited on catalytic and uncatalyzed filters when the feed gas contained 1000 ppm of NO in addition to 10 vol % O₂ in N₂. With respect to the TPO tests in O₂-N₂ only (Figure 2), the presence of NO enhances the soot catalytic combustion rate. The increase in soot reactivity is stronger at lower temperatures (500-700 K) but is also evident at higher temperatures. Comparing the performances of the catalytic filters in Figures 3 and 4, it is observed that, at lower temperatures, the presence of NO increases soot reactivity more than that of H₂O.

Moreover, at higher temperatures, the promoting effect of NO is more marked than that of H₂O with the unsulfated catalytic filter, as the decrease in the peak temperature of the dX/dt curve with respect to that for O₂-N₂ is about 70 K. In the case of the sulfated filter, the effect of NO is slightly weaker than that found with H₂O, the peak temperature of dX/dt curve being reduced from 880 (Figure 2) to 800 K (Figure 4) with NO and to 780 K with H₂O (Figure 3). As in the case of H₂O addition, the uncatalyzed soot combustion seems not to be influenced by the presence of NO in the feed gas, the dX/dt curve in Figure 4 being practically the same as that in Figure 2.

In Figure 4, the reactor outlet NO concentration (NO_{out}) measured in the TPO tests with either uncatalyzed or catalytic filter is also reported. With the unsulfated catalytic filter, at increasing temperature and carbon conversion, NO_{out} decreases rapidly from the inlet value (about 1000 ppm), reaches a minimum (about 700 ppm at 670 K), and then increases approaching the NO-NO₂ equilibrium concentration curve, which is reported in the same figure for comparison. In the case of the sulfated filter, instead, the NO_{out} profile slightly increases up to about 670 K and then decreases, reaching a minimum at 760 K and, subsequently, approximating the NO-NO₂ equilibrium curve. The maximum decrease in NO in this case is only 160 ppm. With the uncatalyzed filter, NO_{out} shows only a slight decrease (less than 100 ppm), with a minimum value at about 930 K in the range of temperatures where carbon conversion occurs. The latter effect is likely due to NO reduction by carbon since, as reported in the literature,^{25,26} at relatively high temperature, NO can be reduced to N₂ by carbon in the presence of O₂.

The combined effect of NO and H₂O on the activity of the catalytic filter in the oxidation of soot is shown in Figure 5, where the reactivity-temperature profiles obtained in soot TPO tests when the reactor was fed with a mixture containing NO, H₂O, O₂, and N₂ are reported. With respect to the separate addition of NO or H₂O, the injection of both slightly increases carbon reactivity with either the unsulfated or the sulfated filter. Furthermore, the shapes of the soot reactivity profiles in Figure 5 reflect the features of both those in Figure 3 and those in Figure 4.

The NO_{out} profiles measured in the presence of water in the reactant gas do not change significantly with respect to those presented in Figure 4. The only difference is that the NO_{out} minimum value pertaining to the unsulfated filter reaches about 500 ppm.

A feature of soot catalytic combustion with $\text{O}_2\text{-N}_2$ is that the catalyst strongly promotes the production of carbon dioxide with respect to carbon monoxide.⁶ In the present work, we have found a CO_2/CO ratio on the order of 100 for the unsulfated filter. The sulfated filter, in contrast, shows a CO_2/CO ratio on the order of 10. Moreover, the additions of H_2O , of NO , and of both reduce the value of CO_2/CO for both catalytic filters. In particular, the ratio varies in the range of 10–40 for the unsulfated filter and in the range of 2–12 for the sulfated filter.

Discussion

The results obtained with the Cu/V/K/Cl/Ti catalyst and reported above show that the activity of carbon oxidation by O_2 is dramatically reduced by sulfation. This can be seen from the dX/dt profiles in Figure 2 and from the reduction in the CO_2/CO concentration ratio, typically occurring with low-activity catalysts.²³ However, the presence of NO or H_2O or both allows the recovery of most of the original activity (Figure 5).

The marked increase in the catalytic soot oxidation rate in the presence of NO was already observed by us in previous works.^{23,24} In these papers, it was found that NO depletion at temperatures as low as 500 K was due to the presence of catalyst, the role of which consisted in oxidizing NO to NO_2 , which, in turn, oxidizes carbon, thereby producing NO again. These reactions, occurring in addition to the direct soot catalytic oxidation with O_2 , increased the overall soot reactivity. Therefore, because the NO_{out} profile was the result of competitive reactions of consumption (NO oxidation to NO_2) and formation (NO_2 reduction with carbon), the observed depletion of NO during the test was smaller when carbon was also present. Moreover, the comparison of the NO_{out} profiles obtained with sulfated and unsulfated catalytic filters (Figure 4) shows that the activity of NO oxidation to NO_2 is also strongly reduced by sulfation.

Whereas the role of NO in enhancing the rate of catalytic soot oxidation is quite clear, the mechanism by which H_2O promotes the reaction is less evident. In the case of the sulfated filter, it could be argued that the increase in catalyst activity observed upon water addition to the feed stream might be related to the decomposition of adsorbed sulfated species. Nonetheless, the results of three consecutive TPO runs, carried out with sulfated catalyst and with water in the feed stream but without the performance of catalyst sulfation after each run, seem to be in contrast with this hypothesis, as the soot reactivity profile does not change. In the literature, the effect of H_2O on carbon black oxidation with a Pt/SiO_2 catalyst²² and on soot oxidation with a $\text{Pt/Al}_2\text{O}_3$ catalyst¹⁷ has been investigated, and the reported results are in agreement with many of the findings of the present paper. Indeed, it has been found that the presence of either NO or H_2O increased the rate of carbon catalytic oxidation, but the role played by NO with such a catalyst was dominant.^{17,22} In fact, whereas in the absence of NO , the effect of H_2O and SO_2 in increasing carbon reactivity with respect to that in the uncatalyzed combustion was relatively small, when NO was present in the feed gas, the addition of H_2O and

SO_2 resulted in a further increase in carbon reactivity. Therefore, in such a system, H_2O and SO_2 have an ancillary role, mainly consisting in making the carbon surface more reactive to the NO_2 attack through the decomposition of relatively stable intermediates.²² A different interpretation was claimed by Cooper and co-workers, who invoked the formation of nitric acid from NO_2 and H_2O , which could act as the oxidizing agent for the carbon surface.¹⁴ In the present work, a significantly different result has been evidenced, as the role played by H_2O is not subsidiary but is as primary as that of NO in enhancing the performance of the catalytic filter. This is clear from the comparison of results reported in Figures 2–4. With respect to the reactivity in the $\text{O}_2\text{-N}_2$ flow, the increase in carbon reactivity upon H_2O addition (Figure 3) is at least comparable to that obtained by addition of NO (Figure 4). Moreover, with the sulfated filter, the soot reactivity peak temperature is lower in the presence of H_2O than in the presence of NO , whereas the opposite occurs in the case of the unsulfated filter.

Altogether, the results indicate that our catalyst promotes the oxidation of carbon at low temperatures in different ways, i.e., by catalyzing both NO oxidation to NO_2 and soot oxidation by O_2 . On the other hand, the EDAX analysis shows that the effect of severe sulfation is the complete substitution of chlorine with sulfur, while the activity of both the direct carbon catalytic oxidation by O_2 and the indirect oxidation by NO are significantly attenuated upon catalyst sulfation. Moreover, from the comparison of the data in Figures 2 and 3, it appears that the activity of the catalyst in the carbon oxidation by H_2O is not attenuated by the sulfation treatment. Therefore, it seems that the loss of chlorine decreases the capability of the catalyst for producing activated oxygen for soot oxidation or for oxidizing NO to NO_2 but not for activating carbon oxidation with steam. Because it is well-known that alkaline metals are good catalysts for carbon gasification by steam,^{29,30} it can be argued that the promoting effect of H_2O on carbon catalytic combustion is mainly due to the presence of potassium and that the mechanism through which this activity unfolds is that typical of catalytic char gasification. Moreover, it is worth noting that the Cu/V/K/Cl/Ti catalyst exhibits a higher resistance to deactivation caused by sulfur in the exhaust than the Pt/SiO_2 catalyst.²² The Pt -based catalyst in Johnson Matthey's Diesel CRT filter requires ultralow sulfur fuel for proper operation.³¹ Further support for the hypothesis that the promoting effect of H_2O on carbon catalytic combustion could be attributed to the catalyst capability for promoting carbon gasification by steam derives from the results of a TPO test carried out with the unsulfated filter by feeding the reactor with an $\text{H}_2\text{O-N}_2$ mixture. The soot reactivity temperature profile for such a test is reported in Figure 6, together with the profiles relevant to the regeneration of the same catalytic filter performed under the conditions of Figures 2 and 3. It is evident that, even in the absence of oxygen, the catalyst is able to oxidize soot at low temperature. Furthermore, it must be underlined that the carbon oxidation in the $\text{H}_2\text{O-N}_2$ mixture starts at the same temperature as it does in the $\text{O}_2\text{-H}_2\text{O-N}_2$ mixture and, therefore, before that in $\text{O}_2\text{-N}_2$. In addition, the dX/dt profile exhibits a peak at the same temperature as that relevant to the TPO test with the

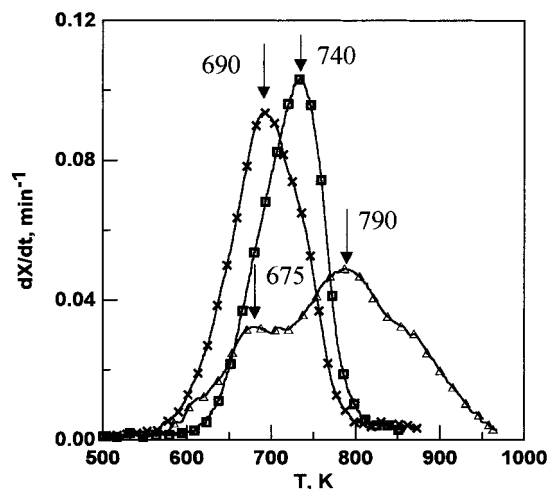
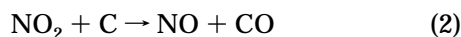
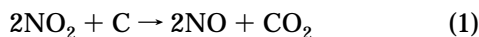


Figure 6. Reactivity-temperature profiles of soot combustion on catalytic filter (CF137TF) in the presence of either O₂ (□) or H₂O (△) or both (×) (feed gas = 0–10 vol % O₂, 0–10 vol % H₂O, balance N₂; gas flow rate = 8.3 cm³/s (NTP); heating rate = 0.17 K/s).

O₂-H₂O-N₂ mixture. Finally, the CO₂/CO ratio found for the H₂O-N₂ mixture is similar to that for O₂-H₂O-N₂.

With respect to the results regarding the selectivity of the carbon oxidation reaction, it was found that catalyst sulfation reduces the selectivity toward CO₂. This can simply be attributable to the decreased oxidation activity of the catalyst after sulfation. However, the addition of H₂O and NO also brings about a reduction in the CO₂/CO ratio, although both species, added separately or together, promote the carbon oxidation activity of the system. These apparently incoherent findings might be due to the particular mechanism through which the oxidation activity unfolds. For instance, it was proposed²⁴ that carbon oxidation by NO₂ and the corresponding reduction of NO₂ to NO might occur through the reactions



where (C-O) represents a carbon-oxygen surface complex. The present results suggest that eq 2 likely plays a major role, giving rise to a net increase in the CO concentration in the reaction products. Similar considerations can be drawn for the addition of H₂O to the reactant mixture.

It is worth noting that, taking into account that both NO and H₂O are present in Diesel exhaust, if their presence results in acceleration of the catalytic combustion of soot, less severe effects of sulfation on the performance of the catalytic filter should be expected in the treatment of a real exhaust. Preliminary results of simultaneous soot loading and regeneration performed at the exhaust of the gas-oil burner confirmed this assumption.³² At 648 K and with the burner operated at an air/fuel mass flow rate ratio $\alpha = 26$, the catalytic filter was continuously regenerated, as shown by the constant value of the filter pressure drop for about 6 h. On the other hand, the planned reduction of sulfur content in Diesel fuel should strongly contribute to a limiting of the negative effects on the activity of soot oxidation catalysts.

Conclusions

The presence of NO and H₂O in the exhaust gas markedly increases the activity of the Cu/V/K/Cl/Ti catalytic filter in soot combustion. The effect of NO can be attributed to carbon oxidation by NO₂, in turn, produced by catalytic oxidation of NO, whereas the effect of H₂O could be attributed to the catalyst capability for promoting carbon gasification by steam.

Severe catalyst sulfation dramatically reduces the activity in O₂-N₂ flow. However, the activity loss caused by the sulfation treatment is partially recovered when NO is present and strongly reduced by the combined effect of NO and H₂O.

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