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INVESTIGATION OF FILTER-CATALYST FOR SOOT EMISSION CONTROL OF OLDER-TYPE DIESEL ENGINES

European standards for diesel particulate matter (DPM) emission impose a ban on further use of old-type diesel engines in Poland and other new member countries of European Union. This particularly refers to engines installed in city buses. Based on the well-known theory that diesel soot particulates have the ability to coagulate and aggregate, we propose a simple and cost-effective method of DPM-emission control. We show the results of successful full-scale tests on DPM emission control system in city buses equipped with old-type diesel engines. DPM emitted from diesel engines is collected on a ceramic filter instead of using a conventional exhaust silencer. A simple automatic regeneration system provides complete combustion of the collected material within 0.5–1 minute. Such a design complies with relevant standards and eliminates potential health implications.

1. INTRODUCTION

Exhaust emission by vehicles with internal combustion engines greatly contributes to the environmental pollution because of still increasing number of vehicles and the distance travelled by each car in year. The pollutants emitted both from spark ignition and diesel engines are divided into two groups: those that are standardized by low-carbon monoxide (CO), hydrocarbons (HC) and nitrogen oxides, and unstandardized species, first of all aldehydes, typical products of incomplete oxidation of hydrocarbon fuels, important due to their potential health effect and their ability to form smog in urban areas. The amount of particulate matter (DPM) in diesel motor exhaust, due to the specific combustion processes in those engines, is also standardized.

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Because of very high emission of air pollutants from spark ignition engines, comprehensive investigations of vehicular emission abatement were concerned with the residual uncombusted or incompletely combusted hydrocarbons, carbon monoxide (a typical product of incomplete oxidation) and nitrogen oxides. Such emission-control devices have been investigated for 30 years which results in the development and standard implementation of catalytic converter into modern gasoline engines, with monolithic three-way catalyst (TWC), that have to meet even more stringent future emission standards. Combustion conditions in diesel engines differ from those in petrol engines: the leaner mixture and higher thermal efficiency make for low HC/CO emissions. The key issue is the emission of DPM. Diesel engines, particularly those for heavy-duty vehicles, with different characteristics of exhaust gas emission demand the other aftertreatment systems to meet more stringently European limits (table 1).

Table 1

The EU ESC emission standards for heavy-duty diesel engines [g/kWh]

	Date of implementation	CO	HC	NO_x	PM	Smoke $[m^{-1}]$
EURO III	October 2000	2.1 5.5*	0.66 0.78*	5.0	0.1 0.16*	0.8
EURO IV	October 2005	1.5 4.0*	0.25	3.5	0.02 0.03*	0.5
EURO V	October 2008	1.5 4.0*	0.25	2.0	0.02 0.03*	0.5

*ETC.
CO – carbon monoxide.
HC – hydrocarbons.
NO_x – nitrogen oxides.
PM – particulate matter.

2. CHARACTERISTICS OF DIESEL PARTICULATE

In table 2, typical emission of pollutants from diesel light-duty engines is compared with emissions of flue gases generated due to combustion of other materials [1].

Burning of wood or coal in small-scale processes and diesel vehicle exhaust are the most common sources of CO, HC and PM emissions, especially in densely populated areas. Nevertheless, PM from diesel engines differs from that produced by fireplace chimneys, and almost all particles of DPM are smaller than 10 μm and are classified as PM-10. It is known that independently of the chemical character, the particles non-toxic in micron-size range may be toxic in nanometre range. While ordinary chimney soot is a carrier of low-boiling hydrocarbons, sulphur compounds and heavy metal oxides adsorbed as a few per cent of a total particle weight, the amount of hazardous substances in DPM may be even as high as 65 wt. % of the total particulate matter. DPM, formed during high-temperature combustion of fuel oil, has a specific surface area of 300 m^2/g and easily adsorbs large amounts of organic and inorganic compounds (figure 1) [2].

Table 2

Typical average emissions due to combustion processes [g/kg of fuel] [1]

Source	CO	HC	NO_x (as NO_2)	SO_x (as SO_2)	Dust, particulates
Power generation:					
gas,	0.1–0.3	0.05–0.08	2–4	0	0
heavy residual oil,	0.5–2	0.2–0.7	5–10	15–30	1
coal	0.1–2	0.03–0.1	1–10	5–20	0.05–2
Municipal waste combustion	0.2–2	0.02–0.1	1–3	0.5–1.5	0.05–0.5
Wood or coal open fires/stove	20–120	2–50	1–5	2–10	1–20
Diesel engine	3–30	0.5–10	5–20	0.5–5	1–10
Otto engine	20–200	10–50	10–60	0.1–1	0.1–0.4
Otto engine + TWC*	2–30	0.5–5	0.2–4	0.1–1	0.05–0.3

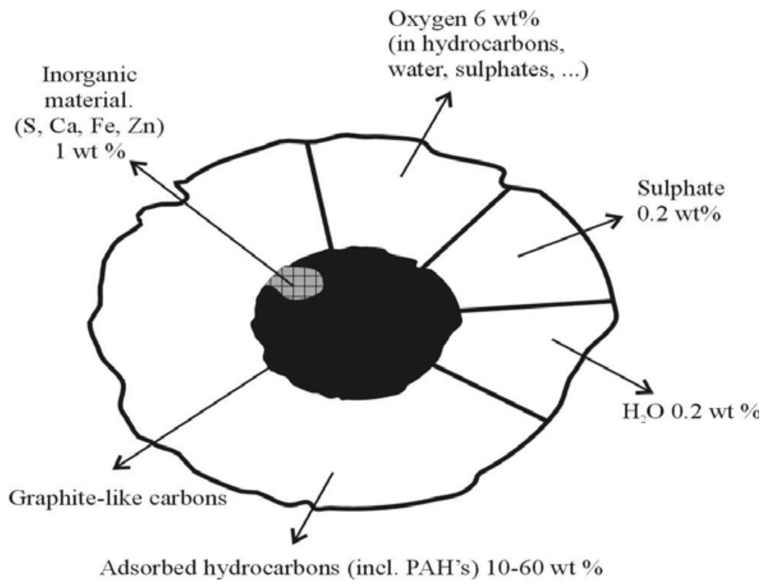

Fig. 1. Chemical composition of a single particle of diesel soot [2]

DPM consists of the soot carbon core, adsorbed volatile and heavy hydrocarbon compounds originating from fuel and lubricant oil (soluble organic fraction – (SOF)), sulphur oxides and metals. Sulphur is present in fuel oil in small amounts (0.001–0.4 wt.%) and is mostly oxidized to sulphur oxides. Small amounts of sulphur are converted into sulphuric acid, which contributes (as sulphates) to the total particulate emission [3]. It has been suggested that a carbon nucleus of the DPM makes a significant contribution to lung cancer induction. Even more dangerous are organic compounds adsorbed on the carbon core, particularly polycyclic aromatic hydrocarbons (PAHs), oxygenated PAHs of high polarity and nitrogenated PAHs. Many of them are recognized as strongly carcinogenic or mutagenic compounds. The composition of volatile organic compounds (VOCs) and PAHs in exhaust gases depends greately not only on combustion conditions but also on the fuel used. The preferred VOCs constituents are methane, acetylene, benzene and ethene, but at lower combustion temperatures there occur unconverted fuel components, aldehydes and α-olefins. The content of emitted PAHs distinctly increased with an increase in fuel aromatization [4].

DPM production heavily depends on motor-operating parameters (such as air to fuel equivalence ratio, volumetric efficiency, ignition start and delay), fuel composition (paraffins, aromatic compounds, additives) and engine design (direct or prechamber injection, injection pressure, turbocharging, etc.). A kinetic scheme of soot, acetylene and benzene production from hydrocarbon fuel proposed by SCHULZ and el. [4] is presented in figure 2.

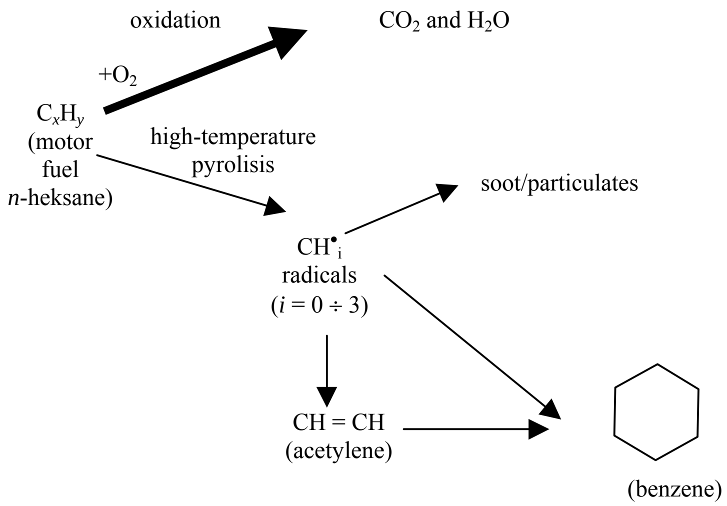

Fig. 2. Kinetic scheme for benzene, acetylene and soot formation in high-temperature pyrolysis during hexadecane combustion in a diesel engine [4]

The fuel is mostly converted into CO_2 and H_2O; some hydrocarbons at high temperatures (>1000 °C) undergo decomposition to C_1 radicals, which can form soot particles or acetylene and eventually benzene. It has been found that emissions are extremely dependent on the air to fuel ratio, and we can conclude that the degree of mixing of the air and fuel at the moment of ignition as well as the combustion temperature are the main process parameters. Diesel soot precursors are transformed chemically and physically in the following processes: nucleation, surface growth, aggregation or chain-forming coagulation and also oxidation (figure 3) [5]. The resulting conglomerates are emitted to the atmosphere as diesel particulates (figure 4) [2].

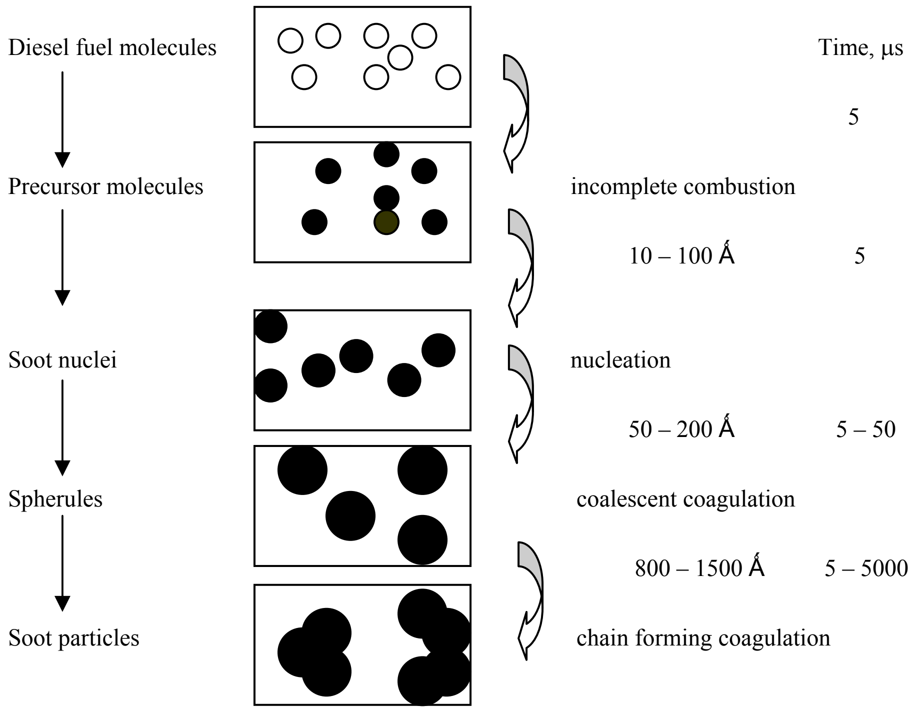

Fig. 3. DPM transformation [5]

The death rate from cancer in cities is generally higher than in rural areas. Epidemiological studies have suggested that long-term employment in jobs with a substantial exposure to diesel exhaust is associated with a 20% to 50% increase in the risk of lung cancer. The risk of lower but more common exposure, e.g., in the urban environment, is much more difficult to assess, but is encountered by a significant part of the human population [1]. The distribution of the particle size by number and mass is shown below [6]:

Particle size	Number [%]	Mass distribution [%]
< 0.1 μm	10.1	0.01
0.1–2.5 μm	89.5	52.6
> 2.5 μm	0.4	47.4

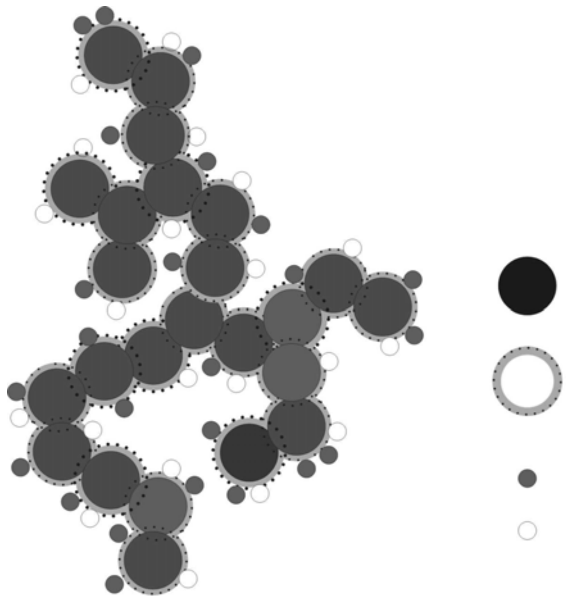

Fig. 4. Composition of the diesel particulate emitted [2]

While a large number (10.1% of the total) of easily inhaled ultra-fine particles contributes very little to their mass, it has been found that equivalent masses of small particles are significantly more inflammatory to the lung than larger-sized particles of a similar composition [6]. Experiments carried out in the 1980s showed that the lowest concentration of DPM responsible for an increased incidence of lung diseases or lung tumors in rats was 50–65 times as high as that in nearby heavy-traffic roads (15 $\mu m/m^3$ per 168-h week) and 6–10 times as high as in workplaces with diesel engines (500 $\mu m/m^3$ per 40-h working week) [7]. DPM emitted into the atmosphere is viewed as a potential carcinogen and it has been considered as such by the International Agency for Research of Cancer (IARC). In 1990, DPM in Germany was officially considered to be carcinogenic and the corresponding threshold value (0.6 mg/m^3 and 0.2 mg/m^3 for underground non-coal mining and for surface workplaces, respectively) was published one year later [8]. It is estimated that DPM emission in the heavy-traffic urban areas is likely to be responsible for thousands of deaths every year. Besides these hazards, DPM contributes significantly to the soiling of buildings, reduces visibility and releases an unpleasant smell, produced predominantly by the aldehydes present in the diesel exhaust.

3. METHODS OF DPM EMISSION CONTROL

Because of a high combustion efficiency and low fuel consumption, the number of diesel engines, both in light- and heavy-duty vehicles, are still increasing. DPM emission can be controlled in different ways. The primary method modification to engines, e.g., direct fuel injection or turbocharging can dramatically reduce PM emission. DPM emission from modern diesel engines is distinctly lower than that from older ones, and particle distribution is shifted towards smaller diameters. The other method the after-treatment devices includes; different types of soot filters such as ceramic filters filled with foams, candle filters, metal-wool or metal-mesh filters were tested. In the modern diesel heavy-duty engines, a nano-filter of heat-resisting ceramic, with the system of spontaneous or forced burning of the DPM collected, is the standard DPM emission control device. When the pressure drop becomes excessive, the pressure sensor at the gas inlet to the filter automatically switches the filter on regeneration. Additional fuel dosed to cylinders increases the temperature of exhaust gases up to soot ignition point. After some seconds of burning, the filtration cycle begins. For easier filter regeneration, some additives like metal-organic compounds are added to the fuel oil, lowering the soot ignition temperature.

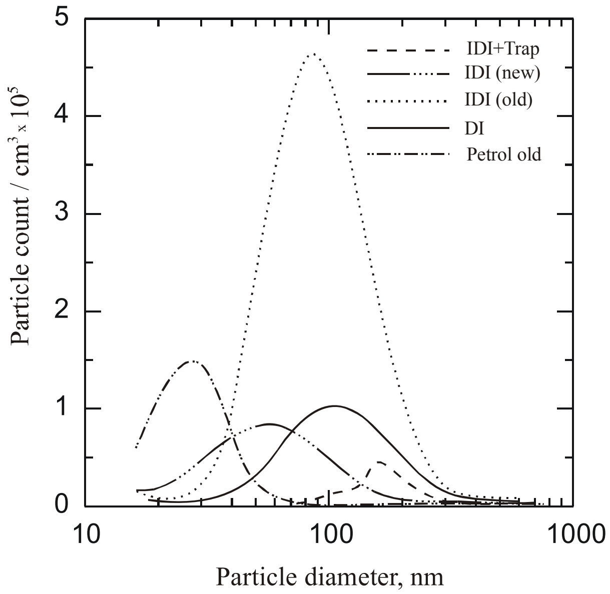

Fig. 5. Particle emission from petrol and diesel engines [9]

Older engines generally emit many more particles with a wide size distribution. Under such conditions, a conventional nano-filter is not efficient; it would be completely blocked in a relatively short time. The highest emissions come from old-type diesel engines, which are still in use in Poland and other countries of Central and Eastern Europe, and will be exploited for at least 5 years. Typical particle-size distribution (by number) for different diesel engine types is given in figure 5 [9].

4. EXPERIMENTS

Our earlier studies carried out with different types of soot filters or filter-catalysts showed that filters used were blocked with soot after relatively short time, and spontaneous filter regeneration by burning out the organic material collected involved very high temperatures, shortening dramatically a lifetime of such filters.

Our recent studies concerned with Ikarus city bus (still very popular in Poland and other Countries of Central Europe) DPM emission control showed that this problem could be solved in a different way. Based on a natural tendency of diesel soot particles to agglomerate, the principle of our method was to increase the diameter of the particles from nanometers to millimeters and to extend an effective filtration time. The size of soot particles can be increased by:

- extending the time necessary for DPM agglomeration,
- installing special partitions in the exhaust flow pathway, which facilitate particle aggregation, and on which the particles can be periodically adsorbed and desorbed, as their diameters become adequately large. Such a partition system enables both an increase in particle diameter and a shift (displacement) in size distribution to above 50 μm.

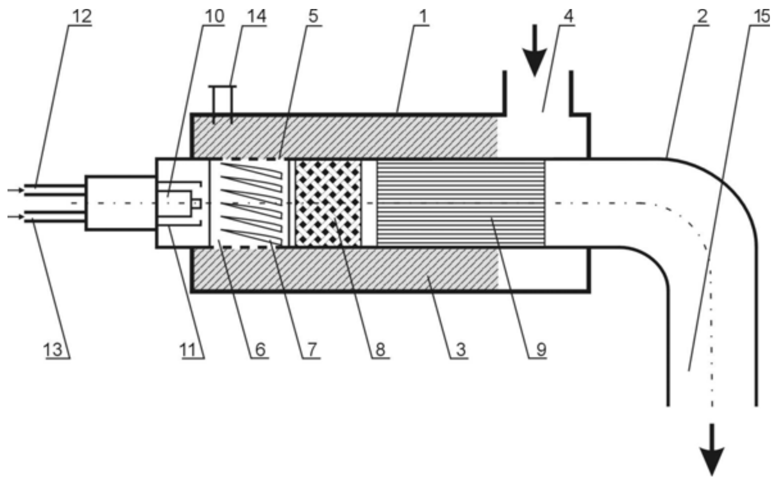

Fig. 6. Filter-catalyst with regeneration in the exhaust gases:
1 – collecting pipe, 2 – exhaust pipe, 3 – porous bed, 4 – inlet,

5 – inlet of exhaust gas to the exhaust pipe, 6 – entry whirl, 7 – skew slot,
8 – filter, 9 – catalyst, 10 – oil burner, 11 – spark igniter, 12 – compressed air pipe,
13 – fuel supply pipe, 14 – pressure drop sensor, 15 – outlet of exhaust gases [9]

This can be achieved in a simple and cost-effective way: the filter must be mounted at the end of the filtration system so that the gas before reaching it first flows through a porous bed of inert material in the collecting pipe [10]. Scheme of the DPM control system is presented in figure 6.

The standard exhaust silencer is replaced with an original filter-catalyst of the same overall dimensions as those of the silencer. The DPM control system consists of:

- a DPM filter,
- a filter regeneration system,
- a catalyst which facilitates the afterburning of CO and hydrocarbons.

In our experiments, use was made of flat, heat-resisting ceramic filters with a thickness ranging from about 2.5 to 4 cm and a pore diameter from 70 to 150 μm. We tested the system with 1 to 3 filters connected in series. The exhaust gases flow through an inert filling, where DPM is aggregated and agglomerated. After attaining an appropriate size, they are entrained by the exhaust gas stream and passed to the filter. The entry whirl produced by skew slot facilitates a uniform adsorption of the DPM on the filter. The duration of filtration cycle depends on the filter type and fuel quality. A pressure sensor is automatically switched on the filter regeneration system. The ignition system of the DPM collected consists of:

- a spark igniter;
- a pump supplying fuel oil to the burner nozzle;
- a system of compressed air supply to the nozzle (to prevent plugging with DPM).

In the filter regeneration system proposed (figure 6), DPM is combusted in fresh air supplied to the burner nozzle together with the fuel; in this time the exhaust gas is not filtered. A monolithic metallic Pt–Pd catalyst used for carbon monoxide and hydrocarbons oxidation is preceded by the system for DPM filtration [11].

The PM emission was measured using optical smokemeter, DO 9500 Radiotechnika, Poland. Based on the emission monitoring under steady-state conditions it was possible to assess the filtration time. Depending on the filter used, an average DPM filtration time – with no negative effect on the engine performance – ranged between 4 and 6 hours. A series of emission measurements were also carried out during tests in city traffic. Hourly measurements revealed a three- to four-fold reduction in emission (table 3).

Recent investigations (from 2004 year) were performed with a reduced space velocity of the exhaust gases (i.e., with a two-fold increase in residence time), which caused the soot to agglomerate, thus to form bigger particles. A series of three 2.5-cm thick filters of cell diameters increased to 1 mm were used. The results revealed a noticeable reduction in DPM emission (figure 7). Such a system enables an extension of an effective filtration time. As for the city buses travelling for 8–12 hours a day, filter regeneration, using burners of special design, can be carried out in the bus depot, and it is not necessary to install the filter regeneration system in every bus. Such a procedure may remarkably reduce the overall costs of DPM emission control.

Table 3

Operation tests of the filter-catalyst mounted in the city bus

Test date	Average smoke emission, [m^{-1}]	Notes
Stationary engine test		
February 14th, 2003	3.13	without particulate filter
	0.83	after 2 hours of idle run
	0.81	after 4 hours of idle run
City traffic test*		
May 10th, 2003	3.10	without particulate filter
	0.80	after 3 hours of operation
	0.77	after 6 hours of operation
*In the course of the city traffic test, spontaneous filter regeneration occurred three times		
City traffic test; carried out on the series of 3 filters with thickness of 2.5 cm (cell diameter of 1 mm) and extending time of soot particle agglomeration		
May 13th, 2004	1.8	without particulate filter
	0.15	after 5 hours of operation
	0.2	after the first regeneration

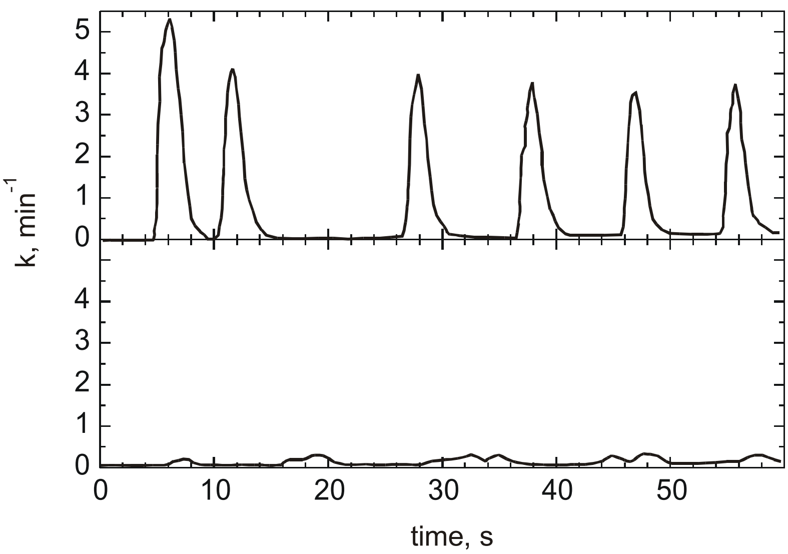

Fig. 7. Results of PM control of a series of 3 filters of 2.5 cm thick
with cell diameter increased to 1 mm; dispersion of smoke particles [*k*]
measured turbidimetrically with optical smokemeter, upper and lower lines – before
and after passing a smoke through a filter, respectively.

4. CONCLUSIONS

To summarize, the design proposed offers the following benefits:

- twice-triple reduction in DPM emissions,
- noise suppression approaches 30% of the noise emitted from conventional silencers,
- installing the system is simple and takes about 1 h,
- the cost is only 2.5 times as high as that of a conventional silencer,
- extending in soot agglomeration time allows DPM to be collected during 8–12 hours and filter to be regenerated in bus depot.

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FILTROKATALIZATOR DO KONTROLI EMISJI SADZY ZE STARYCH TYPÓW SILNIKÓW DIESLA

Zaostrzające się normy emisji sadzy z silników Diesla w Polsce i w innych krajach ostatnio przyjętych do UE mogą stać się przeszkodą dla dalszej eksploatacji autobusów miejskich starego typu. Opierając się na znanej teorii zdolności cząstek sadzy do koagulacji i agregacji, zaproponowano prostą i tanią metodę ich usuwania. Przedstawiono wyniki badań ruchowych filtrokatalizatora zainstalowanego w autobusie miejskim starego typu w miejscu tradycyjnego tłumika. Prosty system automatycznej regeneracji filtra umożliwia wypalenie nagromadzonej sadzy w ciągu 0,5–1 minuty. Zaproponowane rozwiązanie umożliwia dostosowanie wielkości emisji do obowiązujących norm, przyczyniając się tym samym do poprawy jakości środowiska miejskiego.