

PARTICLE FILTER TEST PROCEDURE AND QUALITY STANDARD FOR DIESEL ENGINES

Andreas Mayer

*Technik Thermische Maschinen, TTM, Fohrhölzlistrasse 14 b, 5443 Niederrohrdorf, Switzerland
tel.: +41 56 4966414, fax: +41 056 4966415, e-mail: ttm.a.mayer@bluewin.ch*

Jan Czerwinski

*AFHB/HTI-Bienne, Laboratory for IC-Engines and Exhaust Gas Control
School of Engineering and Information Technology, 2560 Nidau, Switzerland
tel.: +41 32 321 66 80, fax: +41 32 321 66 81, e-mail: jan.czerwinski@bfh.ch*

Marcus Kasper

*Matter Engineering
Bremgarterstrasse 62, 5610 Wohlen, Switzerland
tel.: +41 56 618 66 30, fax: +41 56 618 66 39
e-mail: info@matter-engineering.com*

Gerhard Leutert

*Air Consul, Tavelweg 27, 3006 Bern, Switzerland
tel.: 031 352 30 54
e-mail: gerhard.leutert@gmx.ch*

Norbert Heeb, Andrea Ulrich

*EMPA, CH-8600 Dübendorf, Switzerland
e-mail: norbert.heeb@empa.ch, andrea.ulrich@empa.ch*

Francois Jaussi

*Tecmot, Rue des Grands-Chênes 4, 1752 Villars-sur-Glâne, Switzerland
e-mail: tecmot.f.jaussi@romandie.com*

Abstract

A new approach is needed to test particle filters for retrofitting Diesel engines. Considering the toxicity of the particles as also the physical and chemical attributes of particle filters, the optimal scheme is to test the components themselves independent of the deployment. That scheme ensures the highest effectiveness with least effort. It also enables evaluation of worst-case situations and assesses the hazards of secondary emissions. The Swiss standard SNR 277 205, which mandates the VERT test procedure, is a first step in that direction.

Occupational health specialists have studied the toxicity of ultrafine particles for more than 100 years. Extremely toxic are the particles intruding into the lung. Larger particles, which are always naturally present, are intercepted in the upper respiratory paths. The body then expels these larger particles rapidly via the mucus and cilia. The new manmade particles $< 1 \mu\text{m}$, however massively intrude into the fine pulmonary alveoli, which do not have the cleansing mechanism.

The targeted efficiencies for evaluating modern filters are evident from the progress achieved. Many filters have filtration rates exceeding 99.9%, albeit the directives [4] only require 97%.

Keywords: *combustion engines, diesel engines, particle filters, quality air standards, air pollution*

1. Introduction

Diminishing the emission of solid particles from combustion engines is a primary objective in combating air pollution [1]. The combustion particles also cause global warming more seriously than CO₂ [2]. Improving new vehicles alone cannot yield rapid results. Filter retrofitting is imperative and feasible [3]. The retrofit task confronts a number of challenges. These include: a range of engine sizes (10 – 3,000 kW), heterogeneous engineering designs, widely varying age and operated hours, emission levels, application profiles, fleet composition and infrastructure. The aim is a maximum curtailment of emitted particles with lowest costs and highest reliability. The prerequisite is a new approach to filter testing and certification.

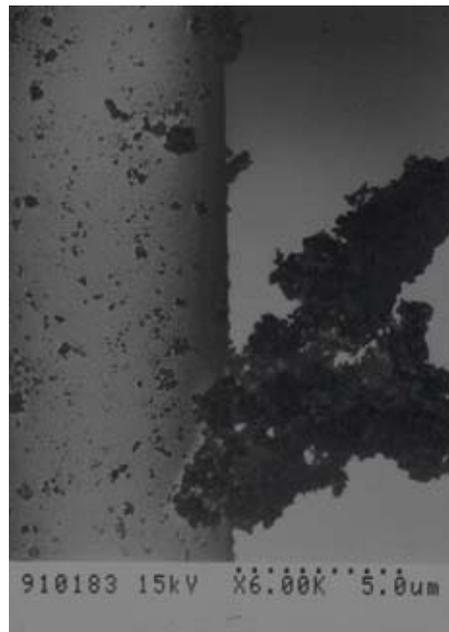


Fig. J. Mayer, ETHZ

The solution is derived from the physics of the filter media. The filtration of fine particles essentially depends on the particle size and the space velocity. The attributes of the emitting engine are only insignificantly relevant. Hence, the physics and chemistry of particle filters can be investigated independent of the engine and its deployment duty. This concept facilitates a very thorough investigation of the size-dependent filtration, aging susceptibility, secondary emissions and extreme situations. Filter systems, which pass this detailed test, perform equally well in every retrofit configuration. This filter test concept was implemented 1998 in the VERT project and is successful for assessing retrofitting [4]. VERT approved filter systems are already deployed in the Low Emission Zones of Europe, North and South America.

2. Objective

Occupational health specialists have studied the toxicity of ultrafine particles for more than 100 years. Their recommendations were codified in the 1959 Johannesburg Convention [5]. Extremely toxic are the particles intruding into the lung. Larger particles, which are always naturally present, are intercepted in the upper respiratory paths. The body then expels these larger particles rapidly via the mucus and cilia. The new manmade particles $< 1 \mu\text{m}$, however massively intrude into the fine pulmonary alveoli, which do not have the cleansing mechanism. There the ultrafine particles can almost unrestrained penetrate the very thin membranes into the blood circulation. Thus the ultrafine particles are transported throughout the body and can even cross the blood/brain and placenta barriers [1, 7].

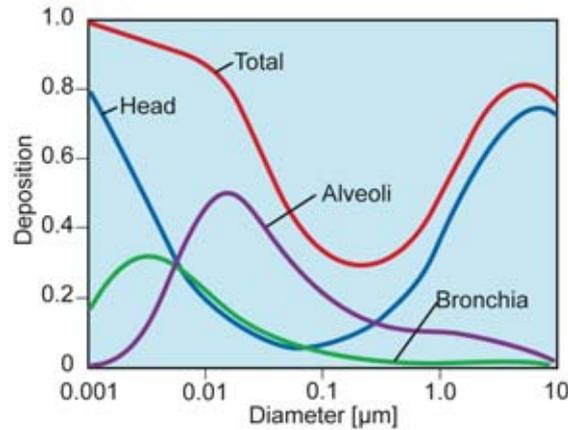


Fig. 1. Fine-particles filtration (as a function of diameter) in the nose, bronchia and alveoli [Source: Helmholtz Institute, Munich]

Consequently, it is crucial to consider the particle size in evaluating particle emissions and the methods to diminish particle emissions. Beside the size, the particle morphology and its solubility are important. Particles, which rapidly dissolve in aqueous ambience, are correspondingly diluted in the body. But insoluble particles retain their toxicity and thus intensify their chronic impact. The morphology is relevant when further toxic substances are deposited on the surface and transported into the body. Soot is mostly inert and insoluble. The deposited substances are mainly PAH (Polycyclic Aromatic Hydrocarbons). This is a “Trojan horse” effect. Both substance categories are classified carcinogenic. Hence, there is no tolerable threshold. The law requires using the Best Available Technology BAT. It is important to distinguish between soluble and insoluble particles, to investigate the existence of accompanying substances and strive for maximum filtration efficacy according to technically rational criteria.

3. Characteristics of particle emissions from utility vehicle engines

Diesel engine emissions of solid particles (“solid” as defined [9] in UN-ECE) display a surprising uniformity (Fig. 2).

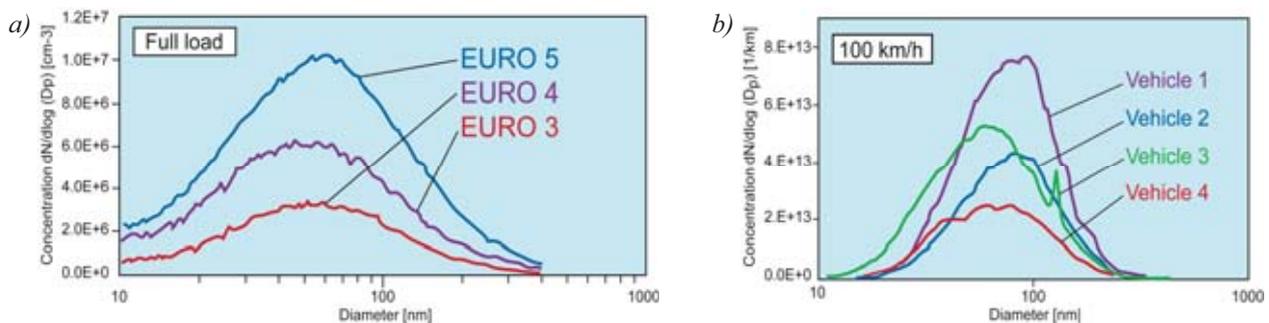


Fig. 2. Particle concentration spectra in the undiluted exhaust-gas: for 3 utility engines (a) [16] and for 4 cars (b) [15]

The plots create the impression that the particles are of different diameters. That is not true. Soot particles are formed as almost same sized “primary particles” [10] of density about 2.4 g/cm^3 and BET surface $150\text{-}200 \text{ m}^2/\text{g}$. The detected larger particles are agglomerates of these primary particles.

After particles enter the atmosphere, dilution prevents further agglomeration. Nevertheless, black smoke is sometimes seen at the tail-pipe. This is because of temporary deposition and aggregation, in the exhaust system, of particles that are subsequently emitted at other operating conditions. This store-and-release behavior predominates in open filter systems [11].

When bimodal distributions of solid particles occur (Fig. 3), then the cause is usually tiny clusters of heavy metal oxides. These originate from lubrication oil or re-nucleation of abraded metal particles [13].

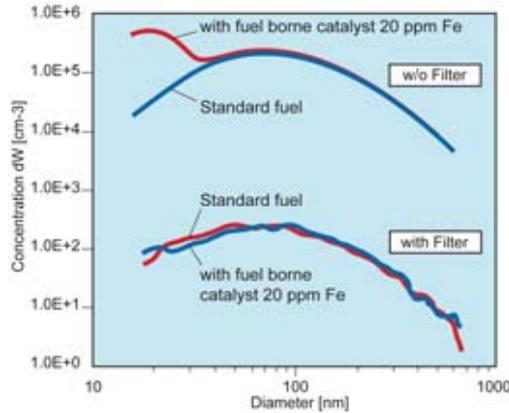


Fig. 3. Bimodal particle distribution, on adding Octimax (a metal-organic fuel-borne catalyst) to standard fuel, with and without filter

4. Physical attributes of soot particle filters

The classic construction of the particle filter is the ceramic cell filter. The exhaust-gas is filtered through the porous walls of cells closed at alternate ends. This concept offers a large filter surface in a compact casing volume. The wall through-flow velocity is a few cm/s . The cell walls have an open pore structure and the pore size is optimally about $10 \mu\text{m}$. Alternative designs are filter membranes of ceramic or metallic fibers, or metallic powders that are sintered to pored structures. Fiber deep filters, electric filter concepts or flow dynamic concepts are yet unsuccessful.

To filter particles in the given size range, deflection effects are ineffective. More functional are impaction and diffusion. Those transport the particles from the flow to the filter surface, where the van der Waal forces firmly bind the particles. Figure 4 shows [6] the effects of impaction and diffusion dependent on the through-flow velocity and the particle size. In the size range above 200 nm , impaction dominates the filtration. Diffusion prevails for smaller particles. Inertia causes impaction. Diffusion mainly depends on mobility. The diffusion velocity of a 100 nm particle is only about $30 \mu\text{m/s}$. Hence, during its wall traverse, it only moves about $1 \mu\text{m}$ closer to the cell wall. This explains the importance of low through-flow velocity and fine pored structures. Canal structures, e.g. so-called particle oxidation catalytic converters, cannot therefore attain high filtration efficiency. No matter how fine the pores are, the new filter structure must collect a light soot deposition, before the filtration efficiency achieves high values. Good filters are sooted within a few minutes. Filter media, which do not form a filter cake, require much longer to attain full efficiency (Fig. 5).

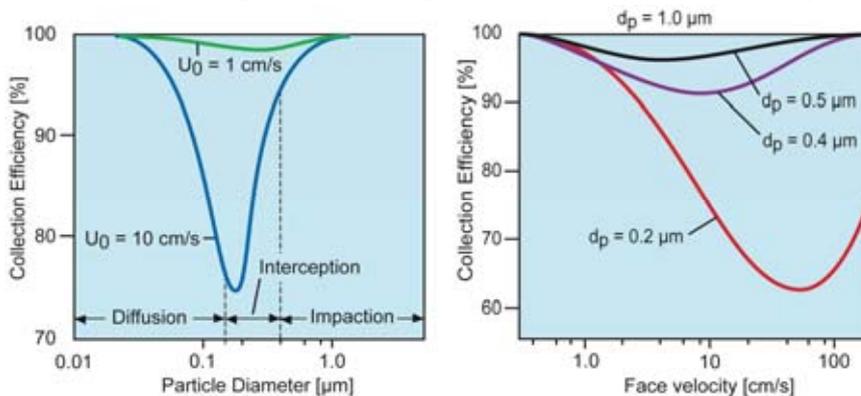


Fig. 4. Filtration efficiency, due to diffusion and impaction, as a function of particle diameter and drag velocity. Hinds [6]

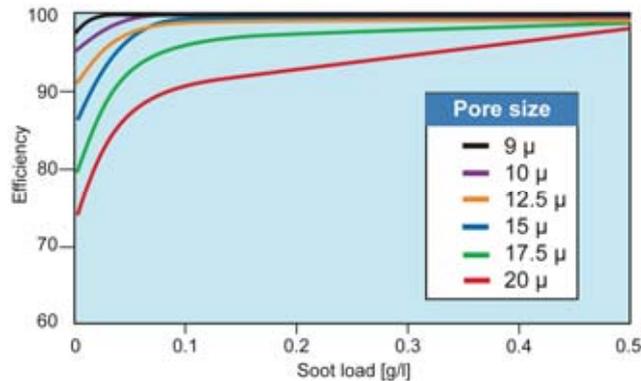


Fig. 5. Filtration efficiency as a function of soot burden and pore size [Source: Haus der Technik "Minimierung der Partikelemissionen" 2005]

Filtration is unsatisfactory when filter cake never forms, i.e. no essentially membrane-like separation occurs but only soot is deposited in the depth of the filter medium. The accumulated soot gradually agglomerates. The consequent flow drag can cause the agglomerate to detach. Hence, deep filters exhibit decreasing filtration efficiency as the soot burden increases. In contrast, wall-flow filters form a filter cake and the filtration rate increases with soot burden [14]. The filtration principle therefore never has a constant response. Filtration efficiency and pressure loss vary with the soot burden; i.e. the response of the burdened and clean filter can be completely different. Hence, the pertinent attributes must be tested. When store-and-release phenomena occur, typically for partial flow filters and so-called open filters, then the response is stochastic and uncontrolled.

The filtration characteristic of soot filters, dependent on particle size, can exhibit completely different trends. See Fig. 6 for four comparative examples.

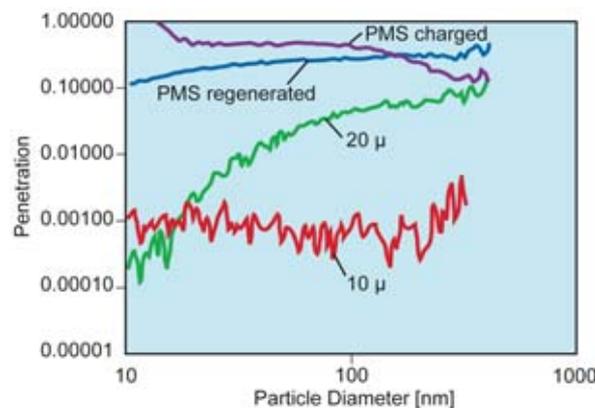


Fig. 6. Penetration characteristic of two-wall-flow filters having pore size 10 μm and 20 μm . Also shown are results for an open filter (PMS) in the regenerated and burdened state [16]. Penetration is defined as the ratio of outlet particles to inlet particles. Penetration is thus $1 - \text{filtration efficiency}$

5. Chemical attributes of filter structures

Filter media must be functional surface-rich structures capable to bind and deposit large quantities of soot (10 g/Liter filter volume). Catalytic effects, due to coating or fuel additives, can transform the filter into a chemical reactor, which synthesizes new compounds from the engine originating gaseous reactants. The NO_x does not nitrate the particle-bound PAH to form highly mutagenous nitro-PAH. However, copper in coatings or fuel additives result in intensive formation of dioxins and furans, and the even minor (10 ppm) chlorine content in the fuel can escalate the quantity of the very toxic substances by 3-4 orders of magnitude [18].

Obviously, the filter is potentially a chemical reactor. Hence, the filter must be carefully tested to ascertain that no toxic compounds are formed. Many countries mandate such verification in the secondary emission tests of the emission legislation [19]. Secondary emissions also include those substances specifically produced to promote filter soot burn-off, e.g. NO₂.

6. Filter regeneration processes

Regeneration designates the continuous or discontinuous, the passive or actively triggered burn-off of the deposited soot [20]. For the emission oriented testing of filter systems, it is important to determine whether during regeneration any reactions occur that increase the emission of toxic substances. The emissions must be tested under realistic regeneration situations, during which the principal toxic components are dynamically measured. There is a European Directive [21], on legislated toxic substances emitted.

7. Metal oxide particles from ash and abrasion

All engines, not only Diesel engines, emit metal oxide particles. These are small, almost insoluble and essentially more toxic than the soot particles of Diesel engines. Metal oxides have a higher surface reactivity [22]. Metal oxides are more toxic than has been recognized in the legislation. To ensure that these pollutants are not emitted at the tail-pipe, the filter test must also include a size-specific trace metal analysis.

8. Fuel consumption

There are consequences if the back-pressure of the particle filter exceeds the pressure loss of the muffler it replaces. The increased pump work diminishes the power output and raises the fuel consumption. The fuel penalty for utility vehicles is below 2%, if the filter is designed compliant with VERT rules, for a back-pressure not exceeding 200 mbar [4]. The influence of the diminished soot emissions is several times that of CO₂ [2]. Hence, the slightly higher CO₂ emissions, due to the filter impedance, are less relevant.

9. Conflict of interests

The objective to minimize particle emissions clashes with practical constraints. These include the space requirement, back-pressure penalty, system complexity, maintenance effort, investments and operating costs. Filters can be made more compact by slightly increasing the pore diameter of the filter walls. Thus at the same pressure loss, a higher space velocity is feasible and the filter becomes smaller and cheaper. Such a filter facilitates impaction; i.e. larger particles are better intercepted. However, smaller particles may escape. Such a filter insufficiency is only detected, during testing, when the filtration efficiency is analyzed size-specific.

10. Unsuitable test concepts

Test concepts based on “Particulate Matter” = PM, i.e. the substance unspecific mass, are misleading. PM contains not only solids, but also condensates including sulphur compounds, which are hygroscopic and bind much water. Thus it is possible that a filter PM is measured, which is greater than the PM without filter [23].

Also unsuitable are evaluations, based on the solid mass, but do not verify the particle size classification.

Testing of dynamic cycles is not recommended. It is extremely complex to dependably measure the size distribution. Filter theory and experience indicate that highly efficient filters are

equally successful in both transient and steady-state driving cycles. This is not true for partial flow filters and so-called open filters, whose structures have a substantially higher flow velocity.

Test concepts having a filter conditioning, before the actual test measurement, are misleading. It conceals both the influence of filter burden on the filtration response, which in reality is an important aspect of filter suitability, and the store-and-release phenomena. Open systems or partial flow filters may have regenerated during conditioning and thus deceptively better than they actually are in realistic deployment.

Very suitable are classification concepts; e.g. those already employed for testing engine inlet air filters, for oil filters, and for cabin filters [24].

11. The test concept SNR 277 205 [20, 25]

In 1993, the occupational health authorities of Switzerland (SUVA), Austria (AUVA) and Germany (TBG) were confronted with a difficult challenge. The WHO had in 1987 specified limiting values for Diesel soot as EC+OC of 200 $\mu\text{g}/\text{m}^3$. This limit was not achievable without very efficient exhaust after-treatment. The occupational hygienists issued an explicit specification. They wanted the almost insoluble particles in the alveoli intruding size range diminished and secondary emissions prevented. That requirement was the impetus for the VERT filter test procedure, which was defined in 1998. After 10 years of successful use of this test procedure, for various retrofit applications, it was defined as Swiss standard SNR 277 205.

Figure 7 describes the multistage test procedure. It comprises a filtration test VFT1, a secondary emission test VSET, a field test VFT2 after 2000 operating hours, and finally an engine rig test VFT3 of the field proven filter. The filtration test is run on a steady-state test cycle, adapted from ISO 8178, at 4 operating points. These are the rated RPM and RPM of highest torque, each at full-load and half-load. Essential is that the operating points, at highest through-flow and maximum exhaust-gas temperature, correspond to the design point of the filter system with maximum space velocity. At each operating point, the concentration of solid particles is determined at minimum 8 logarithmic equidistant size classes between 20 and 300 nm. “Solid” is according to the PMP [9] definition, i.e. a sample heated to 300°C.

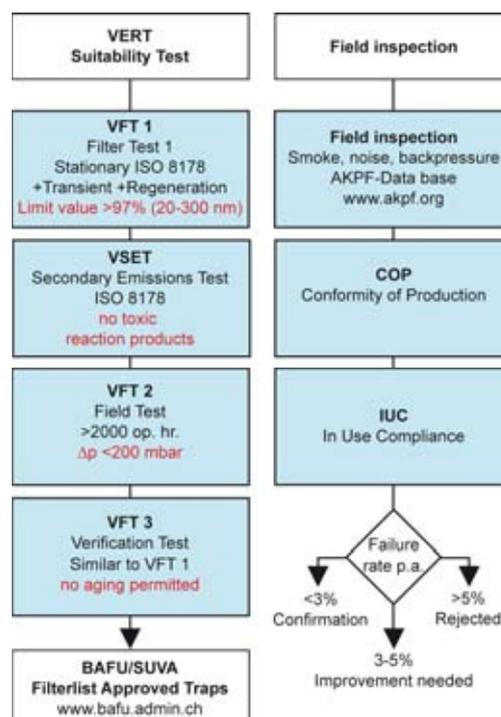


Fig 7. VERT suitability test and field verification

These measurements of the filtration attributes are done both on the new filter, and also on the soot burdened filter before and after regeneration.

The measurements are enhanced with a temperature section to determine the NO₂ formation characteristic. Also done is a test according to the method of free acceleration. This includes a filtration efficiency measurement of the particle count in this transient.

For comparison purposes, the particle mass PM is measured, too, according to the classical method, as well as the occupational health oriented EC carbon mass [27]. Supplementary measurements are the PAS and DC procedures [28]. Those determine the Fuchs surface and precise determination of the fine particle content of the exhaust-gas. Both PAS and DC have transient capabilities. PAS, DC and the size-specific particle analysis SMPS usually correlate very well for evaluating the filtration efficiency. The EC measurement usually shows about 1% worse values, due to the known systematic error of coulometry. The PM based filtration efficiency has a lot of scatter, does not correlate and is not processed in the filter evaluation.

VFT1 also includes the regeneration test. The test determines the risk of toxic emissions during the regeneration. At maximum through-flow, the torque is increased till above the balance point to also investigate the phases of rapid regeneration velocity.

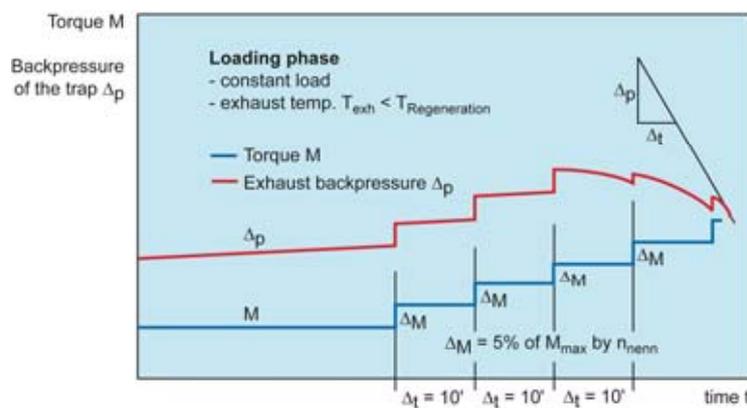


Fig. 8. Regeneration cycle. Plotted are the torque (M) steps and the back-pressure (Δp)

To determine the secondary emissions, the total ISO 8178 C1 cycle is run without interruptions, to collect 200 minutes of exhaust samples, sufficient for detecting trace quantities of very toxic substances. The sample is extracted without dilution from the exhaust-gas, whereby the sample quantity is adjusted to the exhaust gas flow. The exhaust samples are collected in a glass apparatus, which has a cooler, a condensate filtration, a filter and a two-stage absorber (XAD 2) unit [18].

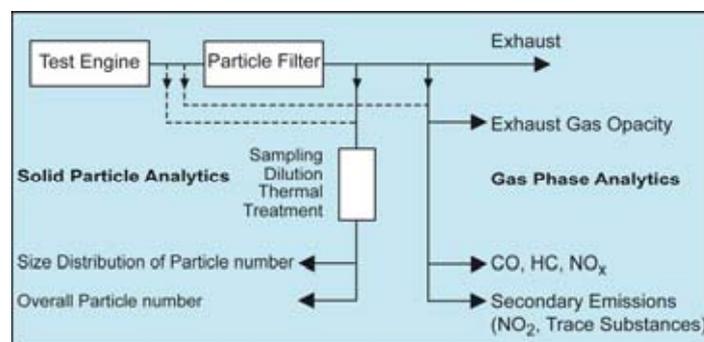


Fig. 9. Sample extraction for secondary components

The sample is analyzed for at least PAH, Nitro-PAH, dioxin and furan contents. Other compounds can be analyzed, too, if formation is suspected [17, 19]. This investigation of the secondary emission not only reveals negative effects. The test also provides much information that

the particle filter not only intercepts solid particle but also efficiently traps 95% of the highly carcinogenic PAH.

The field durability test VFT2 continuously logs the filter back-pressure and the temperature ahead of the filter. The essential evaluation of this test is the analysis of the logged data. That evidences whether the regeneration system performed dependably.

The concluding VFT3 test is essentially the same procedure as VFT1.

12. Benchmarks for evaluating modern particle filters

The targeted efficiencies for evaluating modern filters are evident from the progress achieved. Many filters have filtration rates exceeding 99.9%, albeit the directives [4] only require 97%.

However, not all filters attain this level. Those that only attain 95%, usually suffer store-and-release.

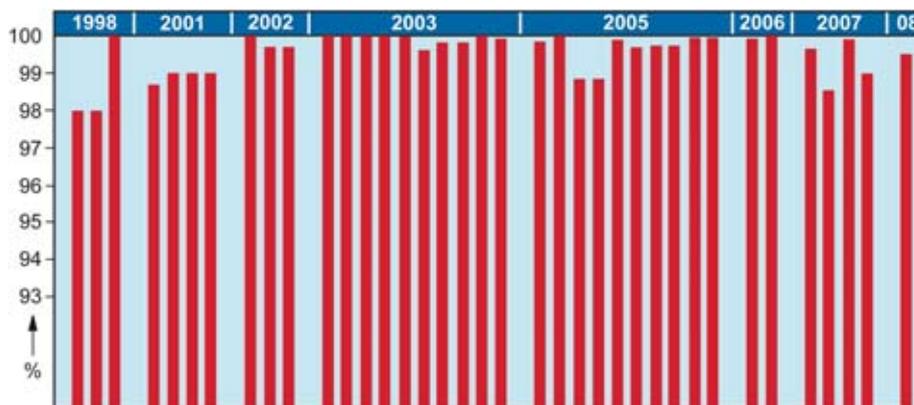


Fig. 10. Results of VERT particle filter tests. Filtration efficiency based on particle count after 2000 hours operation

References

- [1] Wichmann, H. E., et al., *Gesundheitliche Wirkungen von Feinstaub*, ecomed-Verl.-Ges., 2002.
- [2] Jacobson, M. Z., *Control of fossil-fuel particulate black carbon and organic matter*, Journal of Geophysical Research, Vol. 107, 2002.
- [3] Mayer, A., et al., *Reliability of DPF-Systems*, SAE 2004-01-0076, March 2004.
- [4] BAFU/Suva-Filterliste, www.umwelt-schweiz.ch/0607-d.
- [5] Orenstein, A. J., *Proceedings of the Pneumoconiosis Conference*, Johannesburg 1959, London J. & A. Churchill Ltd, 1960.
- [6] Hinds, W. C., *Aerosol Technology*, John Wiley & Sons 1982.
- [7] Oberdörster, *Extrapulmonary effects of inhaled nanosized particles*; 9 ETH-conference, 2005.
- [8] *Air quality guidelines for Europe*, WHO Regional Office for Europe, European Series, No. 23, 1987.
- [9] Kasper, M., *The number concentration of non-volatile particles*, SAE 2004-01-0960.
- [10] Siegmann, K., *Soot Formation in Flames*, Journal of Aerosol Science, Vol. 31, Suppl. 1, pp. 217-218, 2000.
- [11] Andrews, G. E., et al., *Particulate Mass Accumulation and Release*, SAE 2000-01-0508, University of Leeds, Ford Research.
- [12] Czerwinski, J., *Messtechnische Untersuchung offener Partikelminderungssysteme*, im Auftrag des UBA Umweltbundesamtes, www.umweltbundesamt.de/verkehr/techemissmm/technik/pms.htm.
- [13] Israël, G. W., et al., *Analyse der Herkunft und Zusammensetzung der Schwebstaubimmission*, VDI Fortschritt-Berichte, Umwelttechnik, Nr. 92.
- [14] Mayer, A., et al., *Gestrickte Strukturen aus Endlosfasern für die Abgasreinigung, Teil 1*, MTZ Nr. 56, 1995.

- [15] Van Basshuysen, R., Schäfer, F., *Lexikon Motorentchnik*, Vieweg-Verlag.
- [16] Mayer, A., et al., *Nanoparticle-Emission of EURO 4 and EURO 5 HDV*, SAE 2007-01-1112.
- [17] Heeb, N. V., et al., *Secondary Effects of Catalytic Diesel Particulate Filters*, Environmental Science & Technology, Vol. 42, pp. 3773-3779, 2008.
- [18] Heeb, N. V., et al., *Secondary Effects of Catalytic Diesel Particulate Filters: Copper-Induced Formation of PCDD/Fs*, Environmental Science Technology, Vol. 41, pp. 5789-5794, 2007.
- [19] *Section 2002 of the US Clean Air Act*, 6/2002.
- [20] Mayer, A., et al., *Particle Filter Retrofit for all Diesel Engines*, expert-verlag.
- [21] *Uniform Provisions concerning the Approval of Vehicles*, E/ECE/324, Rev. 1/Add 82/Rev. 2/Amend 1, May 2002.
- [22] Risom, L., et al., *Oxidative stress-induced DNA damage by particulate air pollution*, Mutation Research, Vol. 592, pp. 119-137, 2005.
- [23] Mayer, A., et al., *Particle Filter Properties after 2000 hrs Real World Operation*, SAE 2008-01-0332.
- [24] EN 1822: *Classification of HEPA and ULPA filters*, VDI 3677: *Filternde Abscheider*, ISO 5011: *Inlet air cleaning*, ISO/TS 11155-1: *Road vehicle – Air filters*; ISO/TS 13353: *Diesel fuel and petrol filters – Initial efficiency by particle counting*.
- [25] *Testing particle filter systems for combustion engines*, Swiss National Standard SNR 277 205, www.snv.ch.
- [26] *ECE-R24*.
- [27] *VDI-2465*.
- [28] Kasper, M., *Characterization of Nanoparticle Size and Composition*, SAE 2000-01-1998.