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Standard Combustion Aerosol Generator (SCAG) for Calibration Purposes

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Introduction

As solid aerosol, soot particles in exhaust gases of combustion engines are mostly smaller than 1 µm and therefore belong to the finest dust particles in air. Though invisible as single particle, they worsen the air quality in high concentration obviously. Because of their capacity to penetrate into the alveolar region and especially, because of the carcinogenic polycyclic aromatic hydrocarbons (PAH) deposited at their surface, soot particles are considered as an air pollutant with adverse health effects. The controlling of the particulate pollution is of great importance in regard of the varying characteristics of soot particles in dependence of combustion techniques and the exhaust subsequent treatment techniques. In this sense investigations over the physical and chemical as well as the toxicological characteristics of the soot particles must be extended and the particle measuring techniques be improved.

For the research of the characteristics of soot particles as well as for the calibration of the soot particle measuring instruments diesel engines were used up to now frequently as source of soot particles. However, diesel engines indicate several disadvantages for the laboratory investigations of soot particles, such as e.g. the space requirement for a diesel engine is relatively large and the operation of a diesel engine is accompanied from noise and exhaust gas emissions. For the calibration of the soot particle measuring instruments diesel engines are not suitable as source of soot particles, because the size distribution, the number concentration and the chemical composition are only poorly or not at all controllable. Additionally, the characteristics of soot particles at a certain operation point are not steady and reproducible over the entire life span of a diesel engine.

In [1], a graphite generator is described which produces carbon aerosols by spark discharge between particle-delivering graphite electrodes. Though this device is substantially more suitable than a diesel engine regarding the space requirement and the emissions for the laboratory operation, but the produced carbon aerosols deviate considerably from the

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1. C. Helsper, W. Mölter and G. Wenninger, Investigation of a New Aerosol Generator for the Production of Carbon Aggregate Particles, Atmospheric Environment Vol. 27A, No. 8, pp. 1271-1275, 1993

combustion soot particles and are thus in most cases not representative for the latters regarding their chemical and physical characteristics.

The soot generator described in [2] uses a diffusion flame to produce real combustion soot particles. Due to the similar diffusion-conditioned soot formation processes [3], the soot particles from this soot generator must approximate quite the soot particles from diesel engines regarding their chemical and physical characteristics. Since this soot generator forms a particle stream through a steel pipe, which is coaxially put into the top of a diffusion flame. the soot production reacts very sensitively to the modifications of the flow conditions in the steel pipe. Additionally the lower opening of the steel pipe in the flame is inclined to a strong soot deposit.

In the following a new soot generator, called as Standard Combustion Aerosol Generator (SCAG), is presented, which produces real combustion soot particles with selected size distributions in submicron area. The characteristic of the produced soot particles is stable and reproducible respectively.

Principle and Experimental Setup

The principle of the SCAG presented here consists primarily in free-getting soot particles formed in a co-flow diffusion flame of hydrocarbon, by preventing the oxygenation for the flame over a certain flame height. In the next step the particle stream is mixed with quenching gas, in order to prevent further combustion processes in the particle stream and to stabilize soot particles.

The co-flow gas burner consists essentially of two perpendicularly and concentrically arranged steel tubes [Fig. 1]. At their lower end, both tubes are hermetically connected with appropriate gas source in each case. Led out through the middle tube, the gaseous fuel stream is surrounded by a second current consisting of synthetic air, which is led in the same direction to the fuel gas from the gap of the two tubes. In order to produce soot particles, the symmetrical diffusion flame is wrapped with a, tapering upward, circular truncated cone of steel, so that a combustion chamber is formed with an opening at the upper end of the truncated cone as output for the escaping exhaust gases of the flame. The lower end of the combustion chamber was hermetically connected by a seal with the outer tube of the burner. Thus a position for the truncated cone can be found, whereby a smoke column withdraws from the upper opening of the combustion chamber despite burning flame. Even though the oxygen in air coming through the gap is sufficient for the maintenance of the flame, it is but exhausted after a certain flame height. The soot particles, formed in diffusion flame, can be no longer completely oxidized because of the oxygen deficiency over this height and leave the flame by the upper opening of the combustion chamber, nevertheless still wrapped by exhaust gases, which develop around the flame and which prevent particle stream from depositing at devices. The particle stream formed in the combustion chamber is directly initiated into a pipe which is arranged right-angled to the flame axis and hermetically connected with the upper opening of the combustion chamber. In the pipe, an inert gas, e.g. N_2, is supplied to the particle stream in order to quench further combustion processes and to stabilize the soot

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2. L. Jing, Generation of Combustion Soot Particles for Calibration Purposes, Second International ETH-Workshop on Nanoparticle Measurement, 7. August 1998
3. L. Jing, Charakterisierung der dieselmotorischen Partikelemission, Dissertation 1997, Departement für Chemie und Biochemie der Universität Bern

particles. Around this pipe a larger pipe is coaxially attached which is connected with synthetic air source. To dilute the particle stream, synthetic air is supplied to the particle stream through the gap between the both pipes.

The experimental setup of the SCAG is shown in Fig. 2. Four components, i.e. a gas burner, a combustion chamber, a gas supply unit and a pipe for exhaust gaes, form the most important constituents of the SCAG. All supplied gases are connected with the corresponding gas bottle and adjusted using mass flow controller.

The characteristics of the soot particles is influenced particularly by the formation of the diffusion flame in the combustion chamber. With continuous geometry of the combustion chamber and the burner the following parameters play an important role for the characteristics of the generated soot particles:

1. Flow rate of the gaseous fuel and the air coat
2. Composition of the gases taking part in the combustion
3. Quenching position of the diffusion flame
4. type of the fuel taking part in the combustion

Results and Discussions

Operational characteristics

The production of soot particles with the SCAG presented here is characterized by high yield at soot particles. The reason is the fact that the SCAG-procedure efficiently prevents the oxidation of the soot particles. A large part of the soot particles produced in the diffusion flame leave the diffusion flame without oxidation. Due to the high particle concentration in the particle stream large flow rate of the exhaust gases can be obtained by diluting the particle stream.

Since the combustion chamber is shielded from the environment while using defined gases for the flame, the quenching of the diffusion flame as well as the dilution of the particle stream, the production of soot particles as well as their dilution is independent of the environmental influences, such as the ambient temperature and humidity etc.. This is one of the most important prerequisites for a stable and reproducible soot production.

Besides, no condensation was determined at ambient temperature in the exhaust gases. This is to be considered as an important advantage of the SCAG, because complex conditioning of the exhaust gases can be omitted here. A further advantage of the SCAG shows itself in the fact that during operation no soot deposit is possible in the combustion chamber. Since the particle stream is surrounded by exhaust gases in the combustion chamber, the wall of the combustion chamber does not come into contact with soot particles and remains therefore free from the deposits of soot particles.

Resultate

Morphology of the soot particles

The morphology of the soot particles from SCAG was analyzed with a Transmission Electronic Microscopy (TEM) and an Atomic Force Microscopy (AFM), resp.. In Fig. 3, TEM-images of the analyzed soot particles are shown. The soot agglomerates from SCAG are, regarding their structure, comparable or same as the soot particles from diesel engines. The primary soot particles were investigated under AFM. They are approximately spherical and indicate a diameter between 30 to 50 nm.

Size distribution of soot particles

The size distribution and the concentration of soot particles generated by SCAG are determined using a Scanning Mobility Particle Sizer (SMPS). Since the SMPS used works under a concentration of 10^7 particle/cm^3, gas samples of the diluted particle stream in SCAG are additionally diluted using a dynamic dilution system (Dilution unit MD19-1E, Matter Engineering AG).

Like shown in Fig. 4, the size distribution of the soot particles produced by SCAG can be generated for different average values in the submicron area, by e.g. adjusting the characteristics of the diffusion flame and its quenching position respectively. The average value of the particle size distribution can be varied, like those from diesel engines, from approximately 30 nm to several hundred nanometers.

Since fewer oxidation of the soot particles takes place in the SCAG than in a diesel engine, the particle concentration from SCAG is much higher than the particle concentration in the exhaust gases of diesel engines. The particle size distributions with smaller average value of the particle diameter show higher particle concentration in the opposite to those with larger.

Adjustment of the characteristic of the soot particles

In Fig. 5 size distributions of the soot particles are presented, which were produced by the SCAG with constant quenching position and different dilution conditions of the fuel gas Therein it is to be demonstrated that the size distribution of the soot particles can be determined or changed by the adjustment of the gas composition. A rising dilution of the C_3H_8 by N_2 leads to a shift of the size distribution of soot particles to the smaller diameters. Since several intermediate settings among the represented adjustments still can be realized, the shift of the size distribution of soot particles can be executed almost continuously in submicron area.

The amount as well as the oxygen ratio of the synthetic air used for the diffusion flame play an important role for the soot formation processes in the diffusion flame. The influence of the amount of synthetic supply air on the size distribution of soot particles can be clarified with the operation point D in Fig. 5, because, by the reduction of the amount of air, the size distribution of soot particles was significantly shifted to small diameters.

Stability of the soot production and the characteristic of the soot particles

The stability and the reproducibility of the SCAG is described here by the comparison of the size distributions of soot particles, which were produced at the different time with the same operation point [Fig. 6]. Between measurements in larger time intervals the SCAG and the control system were completely switched off.

Both for short and for large time intervals, no significant modification of the size distribution of soot particles was observed. The size distributions in Fig. 6 show clearly that they remain constant over different time intervals. The uncertainty of measurement was under 2% for the measurements, which were executed in time intervals of minutes and hours repectively, and under 5% for the measurements, which were executed on different days.

Apart from the temporal stability and reproducibility of the soot generation, also the consistency of the size distribution of soot particles was examined when diluting the particle stream. It was found out, that, with sufficient quenching gas amount, the dilution of the particle stream does not have influence on the particle size distribution except the total particle concentration. As shown in Fig. 7, the particle size distributions of some operation points of the SCAG indicate no shifts of the size distribution by the dilution of the particle stream. Thus with the new SCAG, a concentration series of soot particles with the same particle size distribution can be formed for the calibration purposes.

Conclusions

The new SCAG enables a controlled production of the real combustion soot particles which correspond, in the most important physical and chemical characteristics, to those from diesel engines. The size of the produced soot particles can be selected by definition and adjusted continuously within the submicron area. The high yield at soot particles offers the possibility for adapting the concentration of the soot particles in exhaust gases for different applications by dilution. The size distribution of the produced soot particles remains despite the dilution of the particle stream unchanged.

Due to the rigid construction of the SCAG, the clearly defined fuel conditions as well as the low soot deposit in the combustion chamber, the SCAG indicates good stability and long-term reproducibility despite e.g. operational interruptions. Compared with diesel engines as soot generator, the new SCAG permits a simple and flexible operation with few noise emissions. Therefore it is particularly suitable for the research of soot particles and the development of particle measuring techniques. Furthermore the SCAG can be used also for the filter check and for the calibration by particle measuring instruments.

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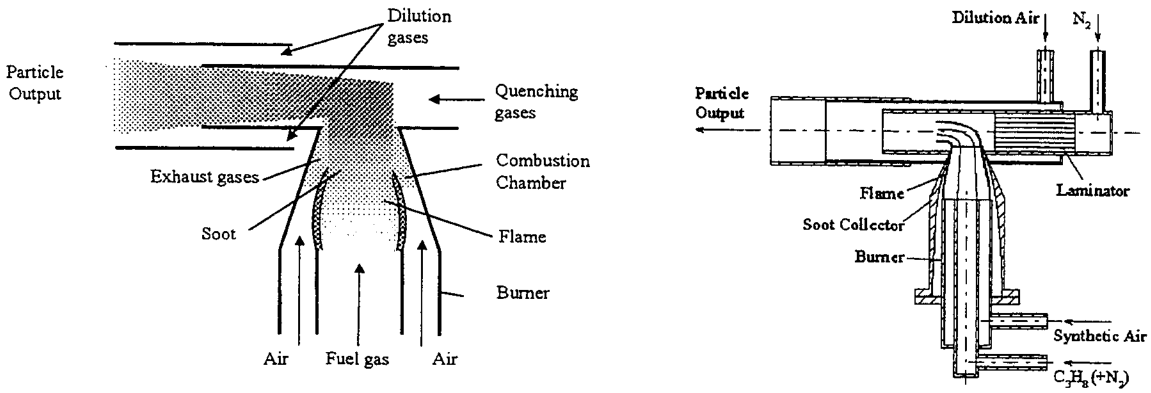

Fig. 1 Operational principle and construction of the Standard Combustion Aerosol generator (SCAG)

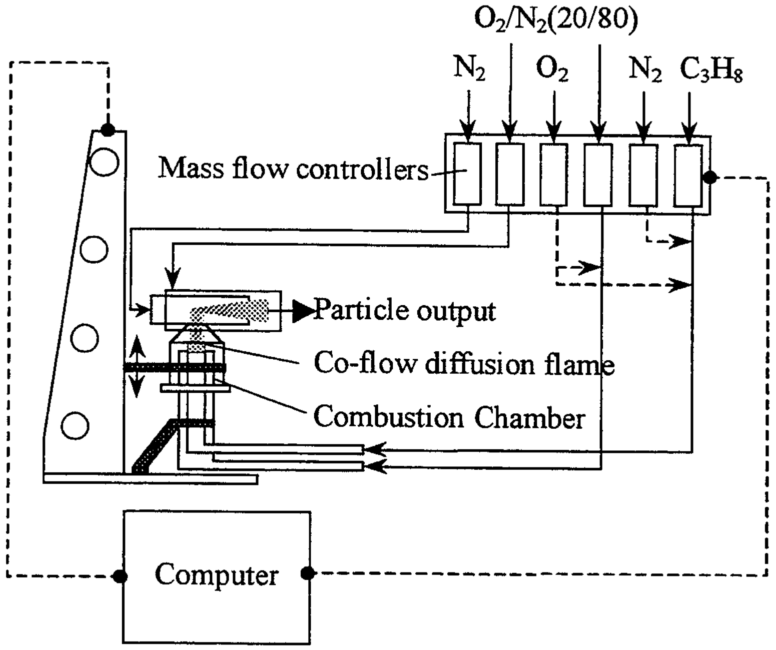

Fig. 2 Experimental Setup of the SCAG

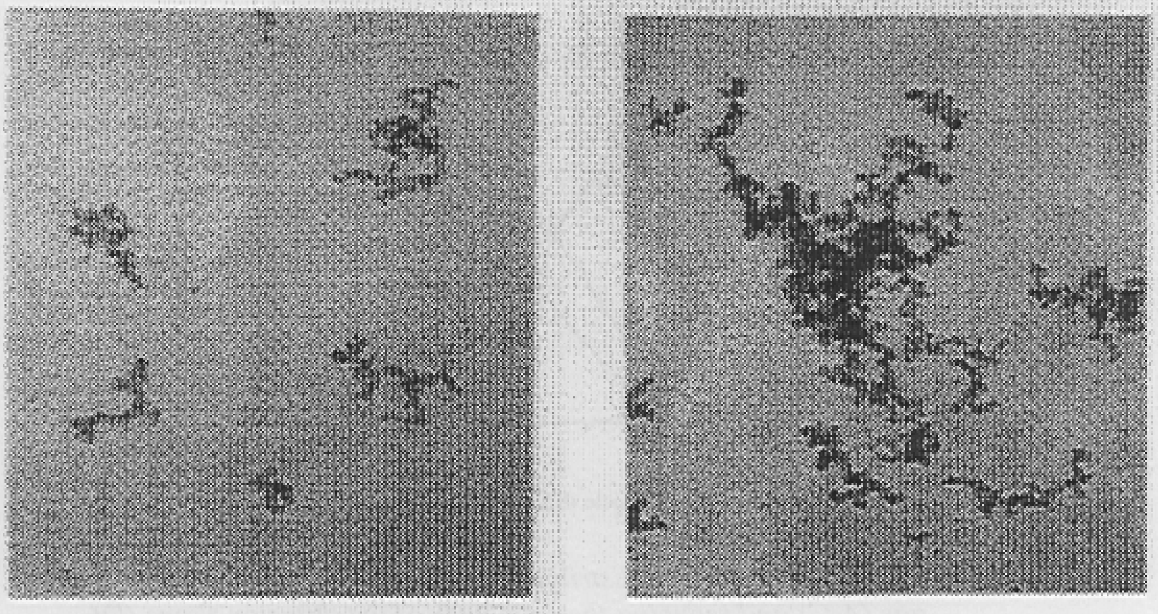

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Fig. 3 TEM-Images of the Soot Particles from SCAG

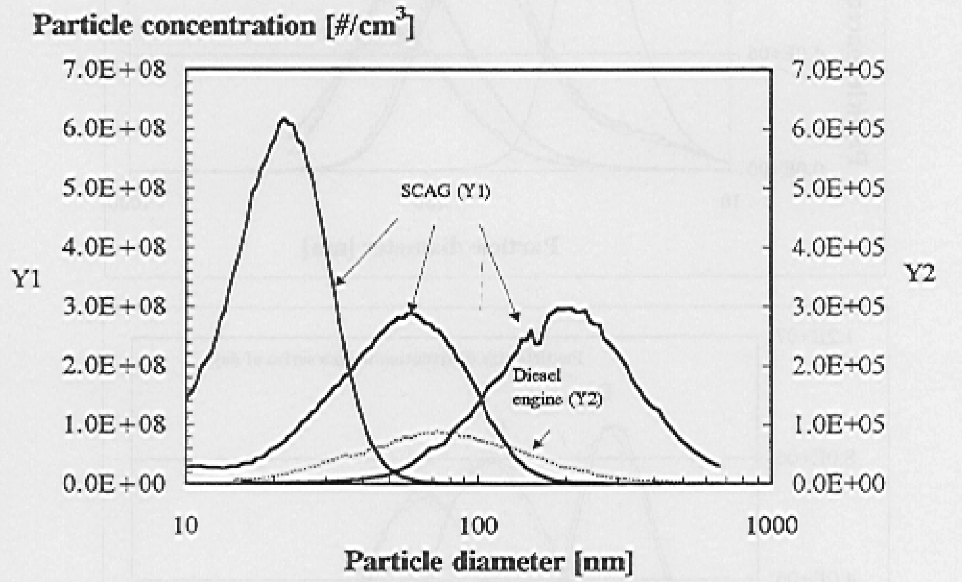


Fig. 4 Different size distributions of soot particles from SCAG, in the comparison with a size distribution of soot particles from a modern diesel engine

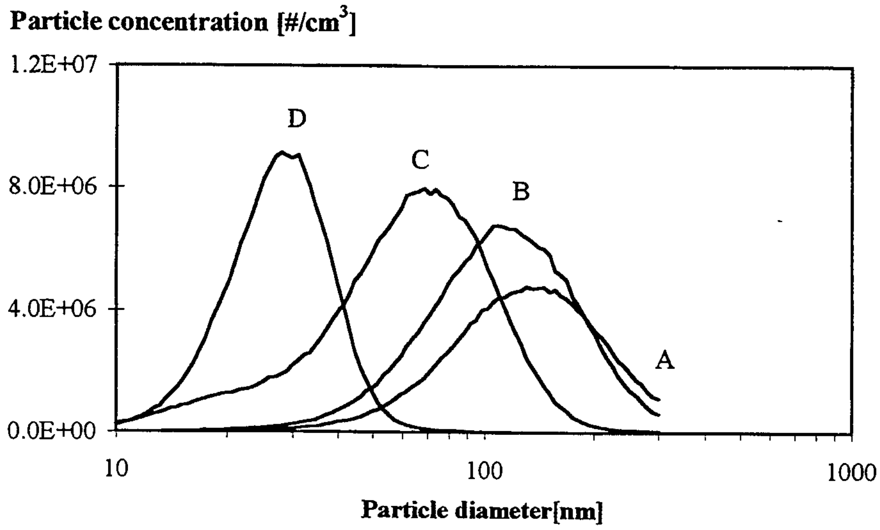
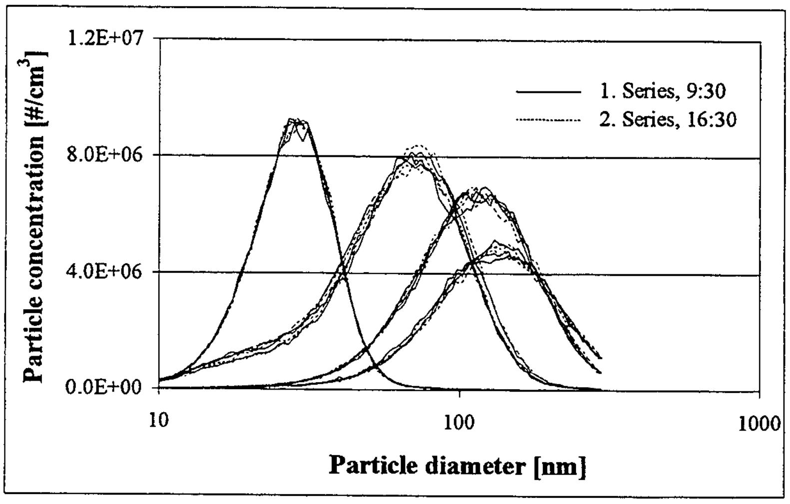


Fig. 5 Adjustment of the size distribution of soot particles by diluting the fuel gas (A-C) and reducing the supply air of the diffusion flame (D)



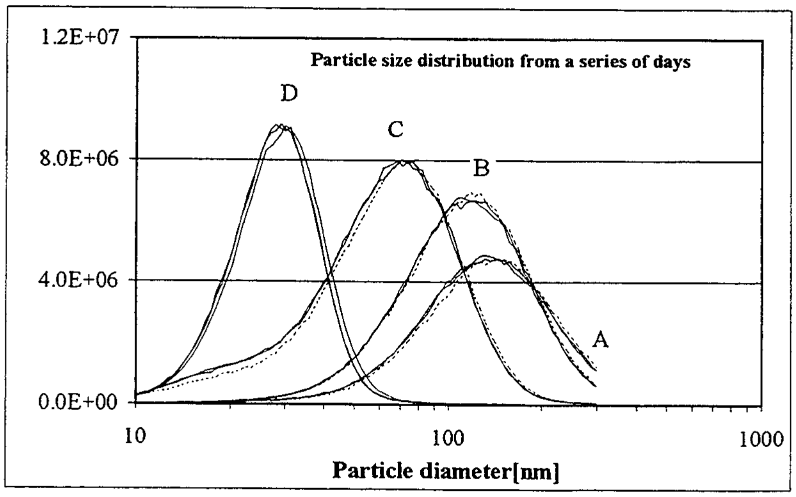


Fig. 6 Stability and reproducibility of the soot particles from SCAG

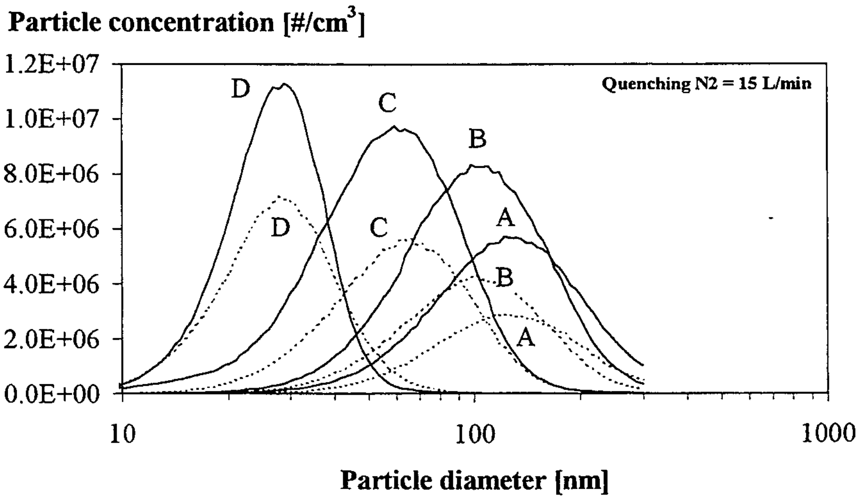


Fig. 7 Size distributions of the soot particles from SCAG before and after the dilution of the particle stream