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Charging-Based PN Sensing of Automotive Exhaust Particles †

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Abstract: Mobile measurement of particle number concentration (PN) in the exhaust of motor vehicles has recently become an integral part of emission legislation. Charge-based sensing techniques for the examination of PN, like Diffusion Charging (DC), represent a promising alternative to condensational particle counters (CPCs) as established PN sensors, because they enable to build robust, compact and energy efficient systems. However, due to the charging process, particle properties like size and morphology have a big impact on the sensor's PN response. For particles of different size and shape we experimentally investigated those impacts using own-built charging-based sensors. The PN response of the DC sensor showed desired behavior for compact NaCl particles, but less satisfying behavior for combustion aerosol standard (CAST) particles, which is a widely used test aerosol for automotive applications. With a photoelectric charger, the PN response of CAST particles was significantly better.

Keywords: particle number; diffusion charging; photoelectric charging

1. Introduction

Examination of PN emissions for type approval of passenger cars is done on test beds according to the particle measurement program (PMP) [1], where a CPC [2] is typically used as PN sensor. However, new regulations require PN measurements apart from laboratory environment, such as on-board determination (known as real driving emissions RDE) [3], or PN monitoring during the vehicle's lifecycle in the course of periodical technical inspections (PTI) [4]. For both methods, simpler exhaust conditioning and more robust, compact and light-weight sensors are needed. For PTI applications there are further requirements with respect to usability and costs. Diffusion chargers can be used for that purpose, however, due to the charging process their PN sensor response significantly depends on the particle size [5]. This requires the modification of the measurement principle by use of a size-selective element. In [5] it was shown that PN determination is possible by means of a diffusion charging instrument based on a modulated precipitation configuration according to the patent [6]. Alternatively to diffusion charging, particles can be charged via the direct photoelectric effect using a light source with sufficient energy to overcome the particle's material work function [7].

2. Theory and Measurement Principle

Conventional unipolar DC is a two-step process consisting of (i) charging of aerosol particles and (ii) subsequent measurement of a current—induced by the charged particles—by means of a Faraday cage electrometer (FCE). In the first step, usually a corona discharge generates positive ions that diffuse onto the particles surface, which imparts a charge on the particles that is, on average,

approximately proportional to the particles size ($\overline{q} \propto d_p$). The measured current I in the second step—given by the product of the flow rate Q, the number concentration N and the average charge per particle $\overline{q}$—therefore also depends on the particle size ($I = N\overline{q}Q \propto Nd_p$). For a PN instrument, the desired sensor response is linearly dependent on concentration only (response $\propto Nd_p^0$). In case of a diffusion charger that is based on induced current measurements [8], the signal's size dependency can be reduced using a pulsed electrostatic precipitator upstream the FCE while operating the charging stage in steady-state mode [6]. The size dependency of the transmission through the precipitation section is opposite to that of the charging process, which enables PN measurements in a size regime relevant for automotive exhaust particles (20–200 nm).

Direct photoelectric charging (PC) is another method by which nanoparticles can be charged efficiently. Exposing the particles to light of low enough wave length (typically in the UV regime) electrons are emitted resulting in a positive net charge of the particles. The PC probability of a particle of charge state q is given by $\alpha^{q \to q+1} \propto K_c I_r (hv - \Phi^{q \to q+1})$ where I_r is the light intensity and hv is the photon energy. Contrary to DC, PC is a material dependent process, which is represented by the material dependent constant K_C, the material work function $\Phi^{q \to q+1}$ (which increases with the particles charging state q), and the material dependent exponent m. For example, the charging probability of NaCl aerosols is substantially lower than for carbonaceous species like soot, which basically enables material specific measurements [9]. The photoelectric charging efficiency is enhanced especially for particles smaller than 50 nm, and it was further shown by Nishida et al. [9] that the sensor response of their photoelectric charger is proportional to the particles surface area ($I \propto Nd_p^2$). In this study, we used a simple photoelectric charger as an additional charging stage within our own-build PN sensor, shown schematically in Figure 1.

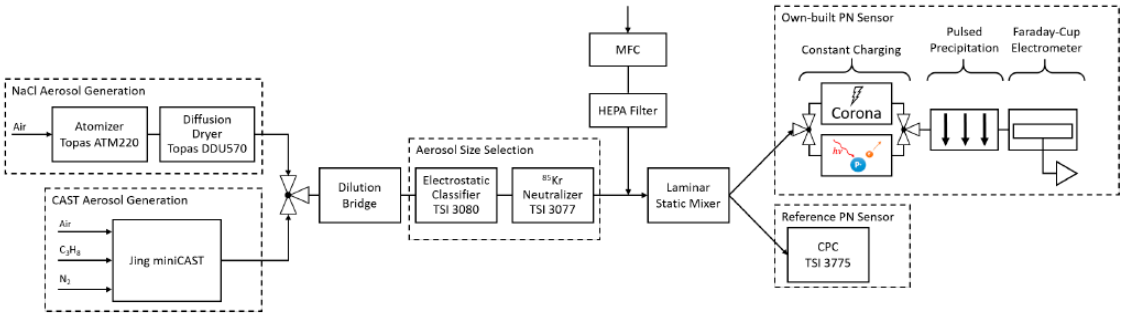

Figure 1. Experimental setup used for sensor efficiency measurements according to [11].

3. Experimental

We used an own-built charging based sensor that works according to the modulated precipitation configuration, whereas either a corona wire charger or a photoelectric charger can be used for steady-state aerosol charging. For the corona charger a thin wire (25 µm) was used as a high voltage electrode (up to 5 kV) to generate positive ions. The ions are sucked through a grounded grid into the charging region where they are mixed with the particles. Excessive ions were captured by an ion trap with an electric potential of −25 V. The photoelectric charging stage consists of a Hamamatsu L7684 xenon flash lamp, which is separated from the charging region by a Heraeus Suprasil 3001 UV transmissive glass disc. The charging region is enclosed by a cylindrical quartz glass tube, surrounded by reflective aluminium. The lamp is operated at a maximum repetition rate of 90 Hz leading to an output power of approximately 4 W.

Within this study we aim to examine the PN response of our sensor, following an experimental setup for the calibration of on-board PN sensors defined by JRC [10]. The setup is shown in Figure 1 and consists of two polydisperse aerosol sources. CAST particles are produced by a Jing MiniCAST 6204 Type B, whereas NaCl particles are generated using a Topas ATM 220 atomizer in combination with a Topas DDU 570 diffusion dryer. A dilution bridge was used to adjust the aerosol concentration before a monodisperse fraction is selected using an electrostatic classifier TSI 3080. The monodisperse

aerosol is further neutralized by a radioactive source TSI 3077 in order to ensure same pre-charging of the aerosols. The monodisperse aerosol stream is further diluted with filtered air and equally distributed using a laminar static mixer, before it is split into our device (operated at a flow rate of 1.5 lpm) and a TSI 3775 CPC as reference instrument (0.3 lpm). With this setup the normalized sensor response of the aerosol charger as a function of particle size is examined by calculating the efficiency for a set of different mobility particle diameters d_m, given by

$$E(d_m) = \frac{I_{Charger}}{N_{CPC} E(70\ \text{nm})}$$

where $I_{Charger}$ is the sensor signal of the aerosol charger and N_{CPC} is the PN reading of the reference CPC.

4. Results and Discussion

The sensor response curves are shown in Figure 2 for two different precipitation voltages of our charging based aerosol sensor. The gray area indicates the limitations for the size dependent sensor efficiency of mobile automotive PN sensors defined by Giechaskiel et al. [11]. For DC, the efficiency curves for NaCl particles stay nicely within these limitations for both precipitation voltages. However, using CAST particles as test aerosol, the size dependency of the efficiency curves is significantly enhanced, especially for dm > 100 nm, causing exceeding of the limits. Because DC is widely known as a material independent process, the different behavior of NaCl and CAST particles may be caused by the different particle morphology. NaCl are compact cubes [12], whereas CAST particles are fractal-shaped aggregates [13]. Morphology effects of DC have been thoroughly investigated within the literature, with no overall agreement. While some studies found a significant increase in the mean charge gained by aggregates [14], other studies only reported a slight difference [15] in charging efficiency between NaCl and soot particles. There may be other effects causing the observed differences apart from the charging process, which is why the reason for the enhanced size dependency of the PN response for CAST particles needs to be studied in more detail by (i) performing experiments with a test aerosol of same material but different morphology, (ii) measurement of the mean charge and charge distribution using various test aerosols and (iii) simulations that account for the particle shape dependent flow behavior. These investigations are ongoing, but are beyond the scope of this work.

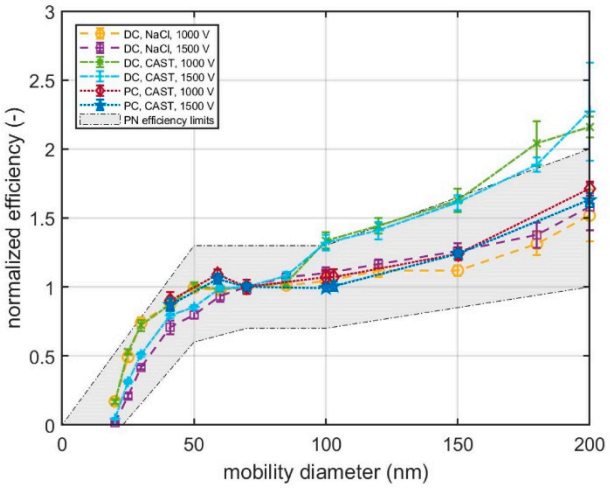

Figure 2. Normalized efficiency curves for CAST (DC + PC) and NaCl particles at precipitation voltages of 1 kV and 1.5 kV.

For PC, where only CAST particles could be used as test aerosol, the sensor response curves clearly remain within the limits for both precipitation voltages. These results are to a certain extend

surprising, since the size dependency of PC was reported to scale with d_p^2 [10]. However, in our PC stage no electric field was used to prevent recombination, which might reduce the size dependency of the charging process. A more detailed characterization of our PC sensor, by means of experiments and a CFD code, is ongoing. Nevertheless, the results show the potential to overcome the morphology problem of modulated precipitation based PN sensors by using PC for the charging process. In recent years UV lamps with sufficient wavelength for PC of soot particles have become available in small frame sizes, enabling their usage within mobile PN measurement systems.

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