The SUREAL-23 project:
Understanding and Measuring Sub-23 nm Particle Emissions from Direct Injection Engines

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Abstract

A large fraction of the total number of particles emitted by direct injection engines are below the adopted 23 nm diameter threshold and although the EU aims to regulate these emissions and impose limits for new light-duty vehicles, this is not yet possible due to the absence of accurate and reliable quantification methods, especially under real driving conditions. The main reason for this is the lack of adequate knowledge regarding the nature of sub-23 nm particles from different engine/fuel combinations under different engine operating conditions. Four research organisations, three particle measurement instrumentation companies and one automotive OEM have joined forces in the framework of the EU-funded project SUREAL-23 to overcome such barriers by introducing novel technology for the measurement of sub-23 nm exhaust particle concentration, size and composition. In this work, we present our latest efforts on advancing particle measurement technology with the introduction of novel techniques and making more robust and the exhaust aerosol sample treatment. Specifically, an induced charged aerosol detector was modified for smaller size and higher temperature particle detection and a differential mobility analyser was adapted for high-resolution particle sizing below 23 nm and high-temperature operation. With respect to sampling/conditioning systems, an advanced dilution system has been developed equipped with a catalytic stripper designed for low particle losses. Additionally, a novel approach has been used with a single hot dilution stage followed by particle measurement systems at higher than the typical temperatures. The understanding of sub-23 nm emissions has been advanced by deploying a variety of fuel-flexible engines and particle generators to produce a wide range of sub-23 nm exhaust particles.

1. Introduction

Vehicles powered by direct injection engines, both Diesel and Gasoline Direct Injection (GDI), are considered a primary source of ambient particle related pollution with impact on climate change (Menon et al., 2002) and severe adverse effects on public health (Alföldy et al., 2009). In June 2012, WHO confirmed the toxicity of diesel particulates and classified them as a “group 1: definitely carcinogenic to human beings” harmful substance. Adverse health effects from vehicle emissions drove many countries, on both sides of the Atlantic, to introduce emission limits. In Europe, emission limits were initially introduced on the basis of emitted particulate mass (Euro 1 to Euro 4). Numerous studies showed that ultrafine particles, with no significant contribution to total emitted mass, are more hazardous on a per mass basis to human health than bigger particles (Ferin et al., 1992) and pushed European legislative authorities to complement the particle emission limits with a solid particle number concentration limit, with a particle size cut-off at 23 nm in Euro 5b for Diesel and in Euro 6 for GDI engines.

Particle number measurements necessitate a robust sampling and measurement method that permits reproducible and comparable experimental results. In Europe, this initiative was led by the Particle Measurement Programme (PMP), which after many years of experimental campaigns and inter-laboratory data analysis proposed a solid particle number measurement method, also known as the PMP protocol (Martini et al., 2009). Accordingly, the raw exhaust is driven to a full flow dilution tunnel utilising a constant volume sampling. Possible volatile and semi-volatile material that condenses on solid particles or creates a separate nucleation mode is evaporated in a volatile particle remover (VPR) and then particle number is measured by a particle number counter having a size cut-off at 23 nm. The VPR consists of two dilution stages: a first stage using hot dilution air and a second with cold dilution air while between these dilution stages an evaporation tube is inserted to bring any already condensed volatile components back to the gas phase.

Fushimi et al. (2011) report that particles in the 10-30 nm size range, regarding composition, may be solid (amorphous or graphitised carbon, metals etc) or liquid (hydrocarbons and sulphur species). According to the insightful review of Giechaskiel and Martini (2014) on engine exhaust sub-23 nm solid particles, the GDI-emitted sub-23 nm solid particle fraction is around 40% while for diesel engines it is 20%. In light of the changing engine technology landscape and the focus on vehicle emissions, mainly due to evolution in engine and exhaust after-treatment technology, the need to measure below the currently enforced 23 nm cut-off in particle size is now well established. One of the problems identified with the standard sampling procedures is the potential formation of particles in the sub-23 nm region, the so-called artefacts, by pyrolysis of hydrocarbon-derived precursors and sulfuric acid re-nucleation (Swanson and Kittelson, 2010). Although the formation mechanism is not entirely understood, the outcome is the production of spurious particle populations that are not present in the exhaust gas.

SUREAL-23 is an EU-funded project that endeavours to investigate in detail the sub-23 nm solid particles emitted by direct injection engines. Starting from October 2016, SUREAL-23 is hosting numerous efforts to:

- Develop new instrumentation to complement standard PMP and extend the available analytical toolset, by providing transient particle number (PN) measurement as well as size and composition classification specifically for the sub-23 nm size region, while pursuing the reduction/elimination of requirements for exhaust sample conditioning by applying high-temperature operation instruments.
- Provide a simple and robust exhaust aerosol sample treatment with increased volatile material removal efficiency and minimal particle losses.
- Investigate the effect of different diesel and gasoline engine operating conditions (fuel additives, bio-content, gas fuel addition, after-treatment type and operation, etc.) on sub-23 nm particle emissions
- Integrate the most suitable components of the extended sub-23 nm measurement toolset proposed developments into Portable Emission Measurement System (PEMS) and verify their measurement capability in real driving conditions.

Herein, we present the up-to-date results of the SUREAL-23 project, including developments in instrumentation, aerosol sampling and conditioning, which have provided some first conclusions and proposals for future investigations.

2. Advanced aerosol measurement technology

1.1. The Advanced Half-mini DMA (HM-DMA)

The HM-DMA system is a supercritical Differential Mobility Analyser (DMA), with a 2 cm working section, initially developed at Yale University and subsequently improved by SEADM, able to classify aerosol particles in the size range 1-30 nm with high resolution (Fernandez de la Mora, 2017A) and fast acquisition frequency (Fernandez de la Mora, 2017B). The working principle involves particle ionisation by a secondary electro-spray and classification under the simultaneous action of the well-controlled axial sheath flow and a strong radial electric field, whereby only the particles of a specific mobility are transmitted to the DMA outlet. A significant feature of the HM-DMA is that it can accommodate hot sampling yielding a device capable of operating at temperatures up to 200°C. By eliminating the need for high sample treatment, known artefact creation mechanisms are avoided resulting in more reliable solid particle emission measurements.

The hot operation of the system was evaluated against sub-23 nm particles generated by a single cylinder, four-stroke, air-cooled and direct injection diesel engine (Hatz, model 1B30) operating at low load (23%) and fueled with high-sulfur diesel (1000ppm S) with addition of 30ml lubricant oil/lt of fuel. Two different experimental setups were used; a single 10-fold hot dilution stage followed by the HM-DMA in hot operation mode and a PMP-compliant setup with a hot dilution stage, a Catalytic Stripper and a cold dilution stage followed again by the HM-DMA for reference measurements. Figure 1 plots the particle size distributions measured with the two setups. The excellent agreement between the two measurements confirms the reliability of the HM-DMA hot operation mode and indicates the possibility of using a simple setup for solid particles measurement.

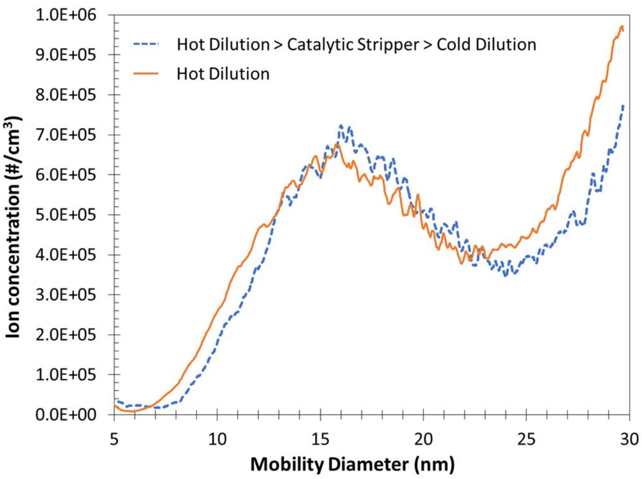

Figure 1. Sub-23nm engine particles size distribution measured with the HM-DMA in hot operation using a PMP-compliant setup and a single step hot dilution.

1.2. The Advanced Induced Charge Aerosol Detector (ICAD)

The ICAD is a concept derived from the induced current measuring principle recently developed by FHNW (Fierz et al., 2014) and marketed as the Automotive Partector by the FHNW spin-off Naneos Particle Solutions GmbH. The evolved automotive-applicable concept involves the addition of a pulsed electrostatic precipitator and closely matches the PN measurement response of the automotive Condensation Particle Counter (CPC) up to 200 nm. The initial design of the device had a cut-off particle size at 23 nm, to be in accordance with the EU legislation. For the scope of the SUREAL-23 project, it was necessary to include the ability to measure smaller particles. Several advances in the design have enhanced its operating temperature range and lowered the cut-off size to around 10 nm. Figure 2 plots the counting efficiency of two advanced ICADs developed in the framework of SUREAL-23. The higher operating temperature range, currently up to 200°C, will allow the use of a much simpler sampling and conditioning system, with lower particle losses, especially in the particle diameter region of 10-23 nm. The small size of the instrument, overall robustness along with the non-demanding sampling system, provide a measurement setup suitable for PEMS.

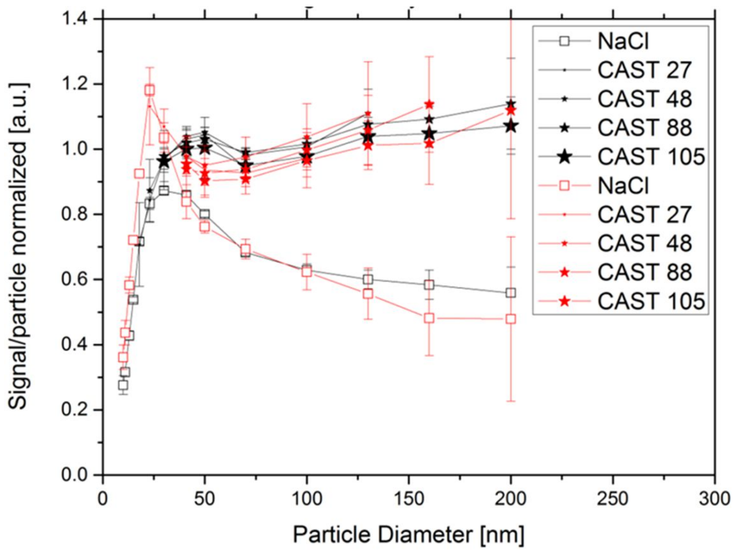

Figure 2. Counting efficiencies of SUREAL-23 advanced ICAD prototypes 2 (black) and 3 (red).

3. Aerosol sample treatment

Within SUREAL-23, a prototype Sampling and Conditioning Particle System (SCPS) was designed and constructed by APTL. Aerosol flow first passes through a hot porous tube diluter and then through a catalytic stripper. A downstream ejector diluter creates suction which drives the sampling flow and provides a further dilution. The whole system is fully integrated and flexible in terms of dilution ratio (DR) and temperature. DR is continuously calculated with a differential pressure measurement across an orifice. Moreover, DR is adjustable and varies in the range 30-120. This is particularly useful for accurately measuring at transient exhaust conditions. Furthermore, the system has been designed so that pressure fluctuation in the exhaust tube does not significantly affect the sampling flow and consequently the dilution ratio. To efficiently remove volatile and semi-volatile compounds without creating artefact particles, the system integrates an in-house developed Catalytic Stripper that oxidises volatile compounds.

DR calculation has shown very good agreement with the CO_2 calculated DR (up to 6% deviation) at steady-state engine points. The results from 20 testing points, where the engine point and the SCPS porous tube and ejector flows and thus the DR were altered. The SCPS was also tested for its tetracontane particle removal efficiency which was higher than 99% for all operating conditions fulfilling the legislation demands. Moreover, solid particle penetration tests showed that the cut-off size d_{50} was at 7.5 nm and goes beyond the current State-of-the-Art. Figure 3 plots the penetration efficiency against the particle mobility diameter.

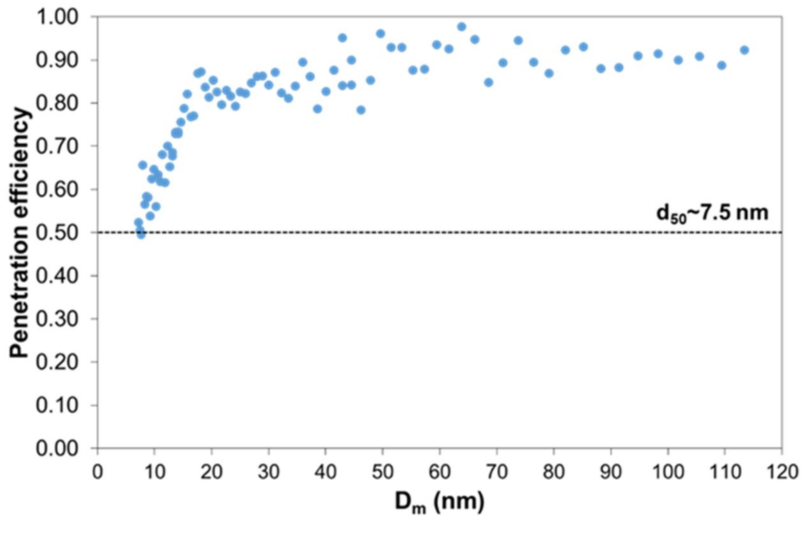

Figure 3. The solid particle penetration efficiency of the prototype Sampling & Conditioning Particle System (SCPS).

4. Conclusions

The objective of our study was to present developments achieved in the EU-funded project SUREAL-23 for the robust measurement of sub-23 nm vehicle emitted particles. We presented two advanced measurement methods: a HM-DMA able to classify exhaust particles with high efficiency in the 1-30 nm size range at elevated temperatures (~200 °C) and an advanced charge-based particle counter (ICAD) with a reduced cut-off size at 15 nm and capable of operation at up to 200 °C. Higher temperature particle instrumentation expands the scope of exhaust particle characterisation while reducing particle losses and sample conditioning requirements. Additionally to instrumentation developments, an integrated exhaust sampling/conditioning system was developed, incorporating a catalytic stripper. The sampling and conditioning system fully oxidises tetracontane particles with size 30 nm while the solid particle penetration cut-off size d_{50} is 7.5 nm.

SUREAL-23 will proceed with further measurement technology developments which will be used for extensive characterisation of tail-pipe out particulate emissions of current and emerging direct injection, internal combustion engine powered vehicles. The effect of different diesel and gasoline engine operating conditions will be elucidated and, more specifically, studies will be performed on the effect of fuel additives, bio-fuel content, gas fuel addition and after-treatment devices on sub-23 nm particle emissions. Finally, the most suitable components of the extended sub-23 nm measurement toolset will be integrated into PEMS and particle emissions under real driving conditions will be examined.

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