Dynamic measurements of soot aggregate size in diesel exhaust by a light scattering method

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Abstract. A light scattering method based on the Rayleigh-Debye approach quantifying the angular scattering from poly-disperse aggregates was tested in an attempt to measure the mean radius of gyration of diesel soot aggregates in diesel exhaust. The aggregate size measured by this method under steady operating conditions showed qualitative agreement with the mode peak mobility diameter obtained by the scanning mobility particle sizer. After the assessment, the method was applied to measure the temporal variation of the mean radius of gyration of diesel soot aggregates under transient mode operation.

1. Introduction
Diesel engines are required to reduce emissions of pollutants including particulates and NOx. In emission measurement protocols in US, EU and Japan the soot particles need to be characterized in mass concentration and size distribution. The diesel particulate in diluted diesel exhaust comprises of elementary carbon particles and secondary particles that are created from volatile hydrocarbons during the dilution process in the air. The measured size of the secondary particle varies significantly with dilution condition, and hence the elementary carbon particle is more reliable to measure.

The size distribution of diesel particles in diluted and undiluted exhaust is generally measured using particle counters such as scanning mobility particle sizer (SMPS) and electrical low-pressure impactor (ELPI). A number of size distribution data published so far revealed that the size distribution of accumulation mode obeys the log normal distribution, and the mode peak diameter varies from 60 nm to 100 nm depending on the engine type and operating conditions. In addition, the standard deviation of the log normal distribution falls within a range 0.2 - 0.3 irrespective of the engine type and operating conditions. Accordingly, the particle size in raw diesel exhaust, which contains no secondary particles, can be quantified only by measuring the average size of the accumulation mode particle.

The diesel particulate filter is made of cordierite or SiC and it is known that the particle filtering efficiency of the filter varies according to the size of particles to be filtered. Thus the particle sizing under transient mode protocol is an important issue in assessing the filter performance.

The present study concerns a trial to measure the average size of accumulation mode particles in diesel exhaust by an optical diagnostics based on the Rayleigh-Debye scattering approach. The optical method tested is expected to have high responsiveness enough to capture the change in particle size under transient mode engine operation.
2. Rayleigh-Debye scattering approach

Diesel soot aggregate comprises a number of primary particles with diameters about 30 nm as shown in Figure 1. The Rayleigh-Debye scattering approach is based on mutual interference between wavelets emanating from different points in an aggregate [1]. Then the size of an aggregate that can be measured by the Rayleigh-Debye approach is represented by the radius of gyration defined by the next equation.

\[ R_g^2 = \frac{\sum_i r_i^2 m_i}{\sum m_i} \]  

(1)

Where \( r_i \) is the distance of \( i^{th} \) primary particle from the center of mass of the aggregate. The size of primary particles is approximately uniform, and the mean square-radius of gyration of poly-disperse aggregates can be written as follows [2].

\[ \overline{R_g^2} = \frac{\sum j R_{g, j}^2 n_j p(n_j)}{\sum n_j p(n_j)} \]  

(2)

Where \( n_j \) is the number of primary particle of \( j^{th} \) aggregate and \( p(n_j) \) is the probability function. When irradiated by a vertically polarized laser beam, a population of poly-disperse soot aggregates present in the exhaust emits scattering with an angular pattern of scattered light intensity according to the size of the mean square-radius of gyration of aggregates. The non-dimensional scattering cross section, \( I^A / n_p^2 \), is given by the Guinier’s law as a function of \( q^2 R_g^2 \) [2].

\[ q^2 \overline{R_g^2} \leq 1.5 D_f \]  

\[ \overline{I^A / n_p^2} = c_1 \exp \left( -c_2 \frac{q^2 \overline{R_g^2}}{3} \right) \]  

(3)

For larger aggregates \( I^A / n_p^2 \) is given by the next equation.

\[ q^2 \overline{R_g^2} \geq 1.5 D_f \]  

\[ \overline{I^A / n_p^2} = \left( \frac{3D_f}{2eq^2 \overline{R_g^2}} \right)^{D_f/2} \]  

(4)

Where \( \overline{n_p} \) is the average number of primary particles per aggregate, \( q \) is given by \( q = 4\pi \sin (\theta / 2) / \lambda \) as a function of wavelength, \( \lambda \), and scattering angle, \( \theta \), on v-v plane. \( D_f \) is the fractal dimension of aggregate. \( I^A \) is the differential scattering cross section of an aggregate normalized by the corresponding Rayleigh cross section for a primary particle and \( \overline{I^A} \) is the mean structural function for differential scattering averaged over all aggregate sizes. Dobbins and Megarisid performed a simulation calculation for forty-five sets of poly-disperse aggregates using \( m=1.57-0.56 \)
and $D_f = 1.7, 1.8$ and $1.9$, and concluded $c_1 = c_2 = 1$ [2]. According to the analysis of transmission electron microscopy images sampled from a 1.7 l diesel engine exhaust, the fractal dimension for diesel soot aggregates ranged from 1.6 to 1.8 [3]. Here, we decided to use $D_f = 1.75$.

The angular scattering cross section, $I^A$, was calculated using equations (3) and (4) against scattering angle with the mean-square root radius of gyration $R_g$ as a parameter. In calculation, $n_p$ for each $R_g$ was estimated using the next equation on an assumption of a 30 nm diameter for primary particle, $d_p$, and pre-factor, $k_f = 8.1$ [4].

$$ n_p = k_f \left( \frac{R_g}{d_p} \right)^{D_f} \tag{5} $$

$I^A$ increases by one order of magnitude when $R_g$ increased from 50 nm to 100 nm because $I^A$ is proportional to the square of $n_p$.

Figure 2 shows curves for $I^A / n_p^2$ versus scattering angle for various sizes for $R_g$, suggesting that the mean radius of gyration is obtainable from measurements of the ratio of the angular scattering intensity at two angles [3]. It should be noted that the angular distribution is relatively flat for small aggregates present near Rayleigh region, while forward scattering dominates for scattering of larger size aggregates. The horizontal line with a height of 0.4166 corresponds to the condition, $q^{2} R_g^{2} = 1.5 D_f$ that links equations (3) and (4). In the domain for equation (4) below the line, it is impossible to obtain $R_g$ from the ratio of scattering intensity at two angles, and accordingly scattering angles of 40 and 70 degree were selected in the measurements of $R_g$ so that at least the scattering intensity at 40 degree stays in the Guinier’s domain above the line even for large aggregates with $R_g$ of 200 nm.

Figure 2. Normalized angular scattering intensity distributions for different mean radius of gyration

Figure 3. Determination of mean radius of gyration from measured ratio of scattering intensities at 40 and 70 degree
Figure 3 shows the relationship between $I^{A}(40)/I^{A}(70)$ and $\bar{R}_g$ calculated with different values of $D_f$. It is seen that the ratio of scattering intensity, $I^{A}(40)/I^{A}(70)$, varies sensitively with $\bar{R}_g$ in the range from 50 nm to 150 nm that includes common size of diesel soot aggregates. It was found that the effect of $D_f$ on measurements of $\bar{R}_g$ is negligible for $\bar{R}_g$ smaller than 130 nm and the sensitivity falls for $\bar{R}_g$ smaller than 50 nm.

3. Optical arrangement

Figure 4 shows a schema of the optical layout for scattering measurements. The exhaust gas sample was lead through a $\phi$ 5 mm x 0.7 m sample line heated at 200 °C to an injection nozzle with 2 mm diameter held vertically. The gas sample was injected upward from the nozzle by the exhaust backpressure into the air in a 24-liter clean chamber. A vertically polarized light beam from a cw green laser (532 nm, 130 mW, Polarization ratio $>100:1$, Laser Quantum-Ventus 532) illuminated soot aggregates in the soot jet at 3 mm above the nozzle exit. The illuminated scattering volume was of a cylinder with a 1.5 mm diameter and 2.0 mm length. The scattered light is collected at a scattering angle by a collecting lens with a focal length of 40.6 mm at a distance of 81 mm from the scattering target. The 15 mm lens aperture defined a solid collection angle of 0.2 sr from the center of the scattering volume. The real image of the whole scattering volume was formed in a plane located at 81 mm from the lens, and the scattered light went through a 3.5 mm x 3.5 mm square aperture in the center of this plane to a dichroic sheet polarizer, a laser line filter (532 nm and 10 nm FWHM) and a silicone photo-diode with a 11.3 mm diameter sensing surface (Hamamatsu photonics S2281-01).

The set of the optics was accommodated in an optical holder that was mounted on a micro optical stage. The vertical and horizontal positions of the holder were carefully adjusted so that the image of the target could be centered in the real image plane in the holder. The detector output was passed thorough an amplifier (Hamamatsu photonics C9329, DC-16 Hz) and was stored on a digital oscilloscope (16 bits, Yokogawa DL 716), sampling at 28 Hz. Since the whole image of the scattering volume was detected at both angles, the correction of the sampling volume length along the laser beam, which is usually made as a function of detecting angle, was not executed. Zero level of the detector output was adjusted by taking the Rayleigh scattering of air molecule, which amounts about 1 mV in this experiment, into account.
4. Calibration of the sensitivity ratio of two light detection systems

Prior to experiments, it is essential to determine the ratio of the sensitivity of the two light detecting systems at two scattering angles. A 2 mm diameter acrylic rod finished with diffused surface was inserted to the jet nozzle to simulate the scattering soot jet irradiated by the laser beam. The scattered light from the rod, whose intensity was different at 40 and 70 degree, was detected simultaneously at the two angles by the two light detecting systems. The sensitivity of the two systems was denoted as $S_1$ and $S_2$, respectively. After acquiring two outputs $D_1 (40)$ and $D_2 (70)$ against light intensity $I_1 (40)$ and $I_2 (70)$ respectively, the light detecting systems were exchanged and another set of outputs $D_1' (40)$ and $D_2' (70)$ against light intensity $I_1' (40)$ and $I_2' (70)$ was measured.

Figure 5 shows the data plots of calibration for determining the ratio of sensitivity. From the two gradient values of the two straight lines, we obtained $S_2 / S_1 = 1.26$ in this calibration.

5. Comparison with SMPS measurements

The reliability of the instrument built here was assessed by a comparative experiment using an SMPS type instrument (Tsukasa, MS-1000). A 3-litre common rail, turbo-charged diesel engine (Isuzu-4JJ1, 2004 year model) was used as a smoke emitter in the experiments. The sample was drawn from the exhaust pipe and lead thorough a 1.5 m flexible stainless tube without thermo-control to MS-1000. The data was acquired simultaneously by the two instruments under various engine torque conditions at an engine speed of 1500 rpm. The soot concentration was changed by changing EGR ratio at each torque condition.

Figure 6 shows the variation of $\overline{R}_g$ with soot concentration, while Figure 7 the mode peak mobility diameter versus soot concentration obtained by MS-1000 measurements. Figure 7 shows a gradual increase in mode peak diameter with soot concentration in high soot concentration range. This is probably due to the small particle loss by the thermophoretic deposition on the low temperature sample line. Except this point, the trend of $\overline{R}_g$ against soot concentration agrees qualitatively with that of MS-1000 measurements, showing a decrease in particle size with soot concentration in low soot concentration range. It should be noticed that the values of $\overline{R}_g$ tends to increase with the increase in engine torque. The reason for this is not clear at the moment but, coagulation of particles might produce larger aggregates in the sampling tube when the exhaust back pressure was increased at high engine torque conditions.

![Figure 6](image6.png)  
Figure 6. $\overline{R}_g$ versus soot mass concentration, showing large values of $\overline{R}_g$ in low soot concentration region (Scattering method)

![Figure 7](image7.png)  
Figure 7. Mode peak mobility diameter versus soot mass concentration, showing smaller change in diameter than in scattering method (MS 1000)
6. $\langle R_g \rangle$ measurements under transient mode

The scattering method was then applied to temporal measurements of $\langle R_g \rangle$ under transient mode operation of the test engine. Figure 8 shows that soot mass concentration measured by the light extinction method in grey line changes according to the change in engine torque [5], while $\langle R_g \rangle$ exhibits opposite trend to soot concentration as expected from the result on Figure 6.

![Figure 8](image_url)

Figure 8. Temporal variations of $\langle R_g \rangle$ and soot mass concentration under transient mode operation

7. Summary

An optical instrument that was based on the Rayleigh-Debye approach was built to measure the mean radius of gyration of diesel soot aggregates in diesel exhaust. The radius of gyration was obtained by providing two scattering intensities detected at two scattering angles into the Guinier aggregate scattering equation. The aggregate size measured under steady operating conditions showed qualitative agreement with the mode peak mobility diameter obtained by the SMPS type instrument. It was demonstrated that the instrument measured successfully the temporal variation of the radius of gyration of soot aggregates in diesel exhaust sampled from a common-rail turbo-charged engine.

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