光学計測によるディーゼル粒子状物質濃度におよぼす諸因子の影響*

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Effects of Various Parameters on Diesel PM Concentration Measured by Optical Method

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ディーゼル機関から排出される粒子状物質(PM)の大きさ、数密度および質量濃度を評価するために Rayleigh DebyeによるClusterモデルによる新しい方法を開発した。実際の測定系は散乱法と透過法を併用したものである。この系によって測定される粒子状物質濃度は粒子状物質自身の物性(光学定数)や使用される光源の波長等が影響因子として考えられる。

本論文では散乱光強度比より決定づけられる凝集体の等体積球相当径(D_a)を固定し、数密度を予測するにあたって、使用光源波長や減衰係数(Q_{ext})の影響が粒子状物質の数密度値におよぼす影響を検討した。さらにレーザー法とフィルター法の両手法の実験結果を論じた。特に低濃度域でのナノ粒子状物質の大きさおよび質量濃度のリアルタイム計測に大きな役割を果たすことが言えた。

Introduction

A number of studies on particulate matters have been established by using either sampling probe methods or optical methods in premixed systems with various types of complex flames, such as real engines and constant volume combustor. Besides, visual inspections of the collected particulate matters were also carried out by electron microscopy analysis by Medalia, 1969; Maraval, 1972; Weralorg, 1972. It seems difficult to obtain ultimate particle sizes and number density due to considerable agglomeration into chains during sampling and along with the traversing time of particulate matters. More recently, laser light scattering was used to make clear diameter and number density of particulate mat-

ters, but the difference between the results of the experiments using laser system and those using filtering system was very large (30%). Christine M. Roussell and Oliver P. et al. used a photographic camera and optical lenses to form a sheet, which was entered into the combustion chamber of a spark ignition engine to get an image in a dense spray and flame body, not particulate matter of exhaust gas. With all the reported methods, particle sizes and concentration profiles in flames may be obtained but not in an exhaust gas flow at condition of atmosphere, where the process of formation of aggregation still happens, consecutively.

The formation and growth of soot particles likely occurs in the central part of fuel-rich pyrolysis. Shapes, sizes and concentration of PM are changing continuously depending on existing and traversing time and many others such as temperature, diameter of exhaust gas tube, etc. Particulate matters in exhaust gas seem to be dispersed or cluster-like,

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spherical or may be in various shapes as shown in figure 1. In order to infer particle size and concentration, the refractive index of PM must be determined as well. If the particles are agglomerated instead of single sphere, thus additional parameters such as extent of agglomeration and particle morphology also need to be inferred. However, it is not easy for determining all the required variables for agglomerated particles, but it can be accurate-

Fig.1 PM Emitted from Diesel Engine

ly formulated, even when an appropriate light scattering for agglomerated particles is employed.

In an effort to measure these parameters, we applied the optical measuring system, which based on the Rayleigh Debye theory of scattering light and attenuation technique to make clear the particle sizes, number density and mass concentration of PM emitted from combustion chamber and diesel engine.

By the Lambert Beer Law, the turbidity of aggregated PM can be certified as follows

$$N_s = \ln(I_0/I_R) (1/L) / ((\pi D_s^2/4) Q_{ext}) \quad \text{.....(1)}$$

Efficiency factor for extinction: $Q_{ext} = Q_{sca} + Q_{abs}$, those Q are calculated by light scattering theory of G.Mie.

Numerical calculation were done to get on overall view on the relationship between the ratio of the two components of scattered light intensity and par-

ticle number in an aggregation N and calculated volume mean diameter D_a with the case of diameter of dispersed particle of 50 nm.²⁾

As particle numbers in aggregation (N) have been changing from 1 to 30 particles, the calculated volume mean diameter of aggregated PM can be certified by the following formula:

$$D_a = D_s \times N^{1/3} \quad \text{.....(2)}$$

On the other hand, according to Lambert-Beer Law, the relation between turbidity τ of aggregated PM in exhaust gas, emerged light intensity I_R and incident light intensity I_0 can be written:

$$\tau = I_R/I_0 = \exp(-\pi D_a^2/4) \times P \times L \times Q_{ext} \quad \text{.....(3)}$$

In formula (3), I_R and I_0 can be measured by experimental photo-multipliers in term of attenuation technique. As a result, number density of aggregated PM can be calculated.

Volumetric fraction:

$$V = -\lambda \ln(I_0/I_R) (1/L) / (6\pi \text{Im}((m^2-1)/(m^2+2))) \quad \text{.....(4)}$$

(m: complex refractive index)

The mass concentration of aggregated PM is obtained be the following equation, as assuming the soot net density 2.0 g/cm³:

$$W = 1.047 \cdot 10^{-15} \cdot D_a^3 \cdot P \quad \text{.....(5)}$$

The experimental results were obtained by means of the light scattering technique through measurements of light scattered from soot contained in exhaust gas of combustion chamber and marine diesel.

This approach, moreover, considers aggregated PM spherical (fig.2) with the diameter of D_a and assumes that mean refraction index of aggregated PM is, $n=1.85$. The value of attenuation efficiency Q_{ext} is shown as in the figure 3 attained by program for calculating Q_{ext} with initial supposed value of volume mean diameter of aggregated PM is from 100

nm to 200 nm. The effects of the optical parameters on the value of mass concentration of aggregated PM were investigated by the cases of attenuation efficiency equals 0.7 and 2.0 and several values of wavelength of incident light for attenuation technique.

Fig.2 Simulation of PM Emitted from Diesel Engine

Fig.3 The Value of Extinction Efficiency against PM Diameter

Experimental Setup

The experiments were based on the light scattering and attenuation techniques. Modulation of the laser light is necessary in order to distinguish the scattered light from the background radiation of the flame and surrounding light, which can cause unexpected signals for scattered light from PM.^{2), 3)}

Incident lights from light sources were chopped by rotating disks ($\phi 10\text{cm}$, 1180 Hz) and concentrated at a point in the exhaust gas flow for the scattering technique or in the form of a pillar band for the attenuation component. The intensities of scattered light from PM have been measured at fixed angular positions of 40° and 140° (angles between the direction of observation and direction of the incident light beam). An optical filter was used to avoid the overload phenomenon of the light intensity signal received in amplifiers. All signals then have been sent to an oscilloscope and computer via analog-digital (A/D) converter, to be processed. The number of signals sent to Lock In Amplifier is one thousand signals, once, to avoid a manual fringe count.

To improve the accuracy of the experiments, all photo-multiplier tubes were put in dark boxes and the equipments were wholly surrounded by dark cloth. Operation of the instruments is primarily automatic, with no user input required once the operating conditions are established. Both hardware and system performance checks are included in the software. This program includes real time displays of the entire size distribution, which are available for automatic control.

The experiments investigating the effects of attenuation efficiency were carried out on the combustion with unsteady flames of $1\text{-}3\text{C}_4\text{H}_6$ gas fuel at supplied pressure of 0.3 kg/cm^2 , the flow rate of air supplied to the combustion chamber was $30\text{ L}_\text{N}/\text{min}$. and the interval of magnetic valve was 0.2 seconds (ON time) and 4 seconds (OFF time). NF-19SK Yanmar diesel engine was also tested using residual oil to sample the particles in experiments investigating the effects of wavelength.

Fig.4 Unsteady Model of PM Generator

Using a grating monochromatic CT-50C (JASCO-Japan Spectroscopic Co., LTD), the wavelength of incident light can be obtained in the range of 340 nm to 550 nm in order to investigate the effects of wavelength to the results of mass concentration.

Fig.5 Schematic of Attenuation Technique with Grating Monochromatic

Fig.6 Engine Setup with Grating Monochromatic

The specification of test engine and fuel oil are shown as in the table 1 and table 2.

Filters were also used to collect the mass concentration of PM emitted from diesel engine in order to compare the PM mass concentration measured by filters to those of PM measured by optical system. The effects of water content in exhausted gas to PM mass concentration measured by

optical and filter methods were investigated by using of two kinds of filters without being dried after collecting particles. The properties of GB-100R and PG-60 filters are shown as in the table 3.

Table 1 Specification of Test Engine

Engine type	Horizontal single cylinder 4-stroke diesel engine
Type and Model	YANMAR NF19-SK
Cylinder Bore x Stroke	110 x 106 mm
Swept volume	1007 cm ³
Maximum power	14 kW/2400 rpm
Rated continuous power	12.0 kW/ 2200 rpm
Cooling type / Weight	Radiator / 192 kg

Table 2 Properties of Heavy Fuel Oil (C oil)

	Unit	Value
Density (15°C)	g/cm ³	0.982
Reaction	-	Neutral
Flash Point	°C	74
Kinetic Viscosity (50°C)	mm ² /sec.(cSt)	2.56
Pour Point	°C	-10
Residual Carbon	mass%	-
S	mass%	0.07
H ₂ O	vol. %	-

Experimental Results and Discussion

1. Results of Experiments with unsteady flames of 1-3C₄H₆ and Q_{ext}=0.7 and Q_{ext}=2.0

Table 3 Properties of Filters

	Unit	GB-100R	PG-60
Weight	g/m ³	95	60
Pressure loss	kPa(mmH ₂ O)	0.3(30)	0.3(30)
Collection Efficiency	0.3 μ mDOP%	99.99	99.9
Heat Resistant Temperature	℃	500	260
Thickness	mm	0.4	0.15
Degree of Stick Water	kPa(mmH ₂ O)	-	7.8(80)<

Example of the measurement is illustrated in fig.7a to 7d in case of Q_{ext}=0.7 of experiments carried out on the combustion chamber with 1-3C₄H₆ at fuel gas flow rate 0.12 L_N/min. and air supplied flow rate 30 L_N/min. Figure 7a shows the profiles of scattering light I₄₀ and I₁₄₀ and the ratio of both intensities I₄₀/I₁₄₀. The scattering light intensities fluctuate complexly with the flow of exhaust gas emitted from combustion chamber. The diameter of soot particles is in the range of 100 nm to 120 nm as indicated in the fig.7b. The number density (N), volumetric fraction (V) and

Fig.7a Light Intensity Signals I₄₀, I₁₄₀ & Ratio I₄₀/I₁₄₀ (1.3C₄H₆, P=0.3 Kg/cm², Q=0.12L_N/min. Q_{Air} supplied=30L_N/min.) ON time: 0.2 sec. OFF time: 4 sec. Q_{ext}=0.7

Fig.7c Light Intensity Signals I₀, I_R & Ratio I_R/I₀ (1.3C₄H₆, P=0.3 Kg/cm², Q=0.12L_N/min. Q_{Air} supplied=30L_N/min.) ON time: 0.2 sec. OFF time: 4 sec. Q_{ext}=0.7

Fig.7b Ratio I₄₀/I₁₄₀ & Volume Mean Diameter (D_a) (1.3C₄H₆, P=0.3 Kg/cm², Q=0.12L_N/min. Q_{Air} supplied=30L_N/min.) ON time: 0.2 sec. OFF time: 4 sec. Q_{ext}=0.7

Fig.7d Number Density, Volumetric Fraction & Mass Concentration (1.3C₄H₆, P=0.3 Kg/cm², Q=0.12L_N/min. Q_{Air} supplied=30L_N/min.) ON time: 0.2 sec. OFF time: 4 sec. Q_{ext}=0.7

mass concentration (W) of aggregated PM can be inferred from the value of mean diameter D_a and the ratio of light intensities of attenuation technique I_0/I_R (fig.7c and 7d). The average volumetric diameter of aggregated PM is around 110 nm and the average mass concentration of PM is about 0.0769 g/m^3 .

For the experiments on the combustion chamber, the optical length $L_1=115 \text{ mm}$.

Figures 8a to 8d display the results of experiments with the same condition with above-mentioned experiments but the value of the attenuation

efficiency is $Q_{\text{ext}}=2.0$. The shapes of the curves for both $Q=0.7$ and $Q=2.0$ are almost the same. The average diameter of soot particles almost do not change but the mass concentration then indicates the average value of 0.0269 g/m^3 which is incredibly different with the result of case of $Q_{\text{ext}}=0.7$.

The comparison of experimental results with various experimental conditions of flow rate of fuel gases and air supplied to the combustion chamber can be observed in the table 4 and figure 9.

2. Results of experiments on 19 PS diesel engine with various values of wavelength of incident

Fig.8a Light Intensity Signals I_{40}, I_{140} & Ratio I_{40}/I_{140}
($1.3\text{C}_4\text{H}_6$, $P=0.3 \text{ Kg/cm}^2$,
 $Q=0.12\text{L}_\text{N}/\text{min}$. $Q_{\text{Air supplied}}=30\text{L}_\text{N}/\text{min}$.)
ON time: 0.2 sec. OFF time: 4 sec. $Q_{\text{ext}}=2.0$

Fig.8c Light Intensity Signals I_0, I_R & Ratio I_R/I_0
($1.3\text{C}_4\text{H}_6$, $P=0.3 \text{ Kg/cm}^2$,
 $Q=0.12\text{L}_\text{N}/\text{min}$. $Q_{\text{Air supplied}}=30\text{L}_\text{N}/\text{min}$.)
ON time: 0.2 sec. OFF time: 4 sec. $Q_{\text{ext}}=2.0$

Fig.8b Ratio I_{40}/I_{140} & Volume Mean Diameter (D_a)
($1.3\text{C}_4\text{H}_6$, $P=0.3 \text{ Kg/cm}^2$,
 $Q=0.12\text{L}_\text{N}/\text{min}$. $Q_{\text{Air supplied}}=30\text{L}_\text{N}/\text{min}$.)
ON time: 0.2 sec. OFF time: 4 sec. $Q_{\text{ext}}=2.0$

Fig.8d Number Density, Volumetric Fraction & Mass Concentration
($1.3\text{C}_4\text{H}_6$, $P=0.3 \text{ Kg/cm}^2$,
 $Q=0.12\text{L}_\text{N}/\text{min}$. $Q_{\text{Air supplied}}=30\text{L}_\text{N}/\text{min}$.)
ON time: 0.2 sec. OFF time: 4 sec. $Q_{\text{ext}}=2.0$

Table 4 Results of Experiments on Unsteady Flames with C₃H₈ and 1.3C₄H₈ Fuel Gases

Cases	Experimental Condition			Experimental Results with Q _{ext} =0.7 (g/m ³)		Experimental Results with Q _{ext} =2.0 (g/m ³)	
	Fuel Gas	Air Supplied					
	Flow-rate (l/s/min.)	Flow-rate (l/s/min.)					
C ₃ H ₈	1	0.2	Natural	0.018	■	0.0063	□
	2	0.3	Natural	0.025	▲	0.0087	△
	3	0.5	30	0.052	●	0.0182	○
1.3C ₄ H ₈	4	0.12	30	0.0769	★	0.0269	☆
	5	0.15	50	0.14	◆	0.049	◇

Fig.9 Results of Experiments on Unsteady Flames with Q_{ewt}=0.7 and Q_{ext}=0.2

light.

The optical length in these experiments was $L_2=40$ mm. The scattering light intensities I_{40} and I_{140} , the ratio I_{40}/I_{140} and the volumetric diameter D_a of aggregated PM are shown in the figures 9a and 9b. Figures 9c and 9d show the light intensities of attenuation technique I_0 and I_R , the ratio I_0/I_R and the distribution of number density N , volumetric fraction V and mass concentration W of aggregated PM exhausted from diesel engine with residual oil at 50% of engine load. These results were obtained when He-Ne laser light source with wavelength of 632.8 nm was used for attenuation technique. The

measurement of PM mass concentration were also done with the cases of 0%, 75%, 50% and 100% of engine load with the same wavelength 632.8 nm. When the engine load increases the mass concentration of PM in 1 m³ of exhaust gas increases and lies in the range of 0.0372 g/m³ to 0.1123 g/m³.

At each of diesel engine load condition, the experiments were also carried out with different wavelength of incident light used for the attenuation technique from 340 nm to 540 nm to measure the mass concentration of PM and the results are shown as in the table 5. The concentrations of aggregated PM were calculated with optical length $L_3=160$ mm.

Figures 11 show how the mass concentration of obtained PM depends on the wavelength of incident light that was used in the measurements. The PM concentration in 1 m³ of exhaust gas emitted from diesel engine increases with an increase in the diesel engine load, but the volumetric diameters of aggregated PM are not very different. Besides, all of the marks, which stand for the results obtained in cases of 632.8 nm of incident light, distributes at the lowest places in the graph. It appears that the mass concentration of aggregated PM obtained by this Rayleigh-Debye method depends slightly on the wavelength of incident light was used for the attenuation technique. It can be observed that at lower incident light wavelength, the aggregated PM becomes higher in concentration. It seems that the shorter the incident wavelength becomes, the higher the occurrence of being obstructed by particles in exhaust flow is.

3. Results of experiments obtained by laser and filtering methods

The comparison of results of experiments obtained by this optical system and those of filtering system imply that the well-designed Rayleigh-Debye measuring system enhances high sensitive of measurement, especially for low PM concentration zones. With almost the same above conditions of experiments, the PM mass concentrations were also collected by GB-100R and PG-60 filters. Figure 12 displays the PM mass concentration collected

Fig.10a Light Intensity Signals I_{40} , I_{140} & Ratio I_{40}/I_{140}

Fig.10c Light Intensity Signals I_0 , I_R & Ratio I_R/I_0

Fig.10b Ratio I_{40} , I_{140} & Volume Mean Diameter (D_a)

Fig.10d Number Density, Volume Fraction and Mass Concentration

Table 5 Results of Experiments with Different Wavelengths of Incident Light for Attenuation Technique

Incident Light Wavelength (nm)	Concentration of Aggregated PM (g/m^3) vs. Engine Load				
	0%	25%	50%	75%	100%
340	0.0396	0.0631	0.0785	0.0102	0.1138
360	0.0391	0.0623	0.0798	0.0908	0.1149
380	0.0377	0.0672	0.0828	0.1005	0.1160
400	0.0389	0.0668	0.0824	0.0953	0.1151
420	0.03865	0.0598	0.0786	0.0966	0.1122
440	0.03851	0.0661	0.0792	0.0982	0.1172
460	0.0383	0.0652	0.0781	0.0976	0.1153
480	0.0384	0.0615	0.0774	0.0922	0.1157
500	0.0382	0.0633	0.0776	0.0956	0.1152
520	0.0381	0.0621	0.0772	0.0932	0.1136
540	0.0376	0.0582	0.0760	0.0911	0.1146
632.8	0.0372	0.0591	0.0769	0.0902	0.1123

by GB-100R and PG-60 filters, which are quite different to each other due to the filter quality but the curve showing the results of optical system lies between both of these curves. It seems that because of higher quality, amount of water collected by GB-100R filters is much more than that of PG-60 filters but this amount of water does not make any differences in values of PM mass concentrations measured by optical system. This explains why the GB-100R filters gave the higher PM mass concentrations than those of PG-60 filters.

Fig.11(a) The Distribution of PM Mass Concentration vs. Engine Load at Different Wavelength of Incident Light

Fig.11(b) The Distribution of PM Mass Concentration vs. Engine Load at Different Wavelength of Incident Light

Fig.12 Comparison of Experimental Results of Laser System and those of GB-100R and PG-60 Filtering System.

Conclusions

For the effectiveness and high sensitiveness of application of Rayleigh-Debye measuring method some suggestions were summarized as conclusions of this study.

- The refractive indices and attenuation efficiency of aggregated PM emitted from marine diesel should be chosen at values of 1.85 and 0.4 to 0.7, respectively.
- Short wavelength of incident light seems to give good results for measurements and the higher the power of incident light sources becomes the higher sensitiveness of measuring system can be obtained.
- The GB-100R filter collects higher amount of PM mass concentration than that of PG-60 in almost conditions of engine loads and the appli-

cation of this Rayleigh-Debye method instead of filters is recommended for measuring PM mass concentration at very low concentration zones.

Nomenclature

- D_a volumetric mean diameter of aggregated PM (nm).
 D_s diameter of dispersed PM (nm).
 I_0 incident light beam intensity (in equal unit of mV).
 I_R emerging light intensity (in equal unit of mV).
 L light path length (cm), $L_1=115$ mm, $L_2=40$ mm, $L_3=160$ mm.
 m_0 refractive index of the surrounding medium.
 N_s segment number of dispersed PM in an aggregation (particles).
 n mean refractive index of aggregated PM.
 P number density of aggregated PM (particle/cm³).
 Q_{ext} extinction efficiency.
 V volumetric fraction of aggregated PM (cm³/cm³).
 W mass fraction of aggregated PM (g/m³).
 λ wavelength of incident light.
 θ angle between scattered and incident light.

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