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PRIMARY AND AGGREGATE SIZE DISTRIBUTIONS OF PM IN TAIL PIPE EMISSIONS FORM DIESEL ENGINES

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Particulate matter (PM) emission exhausted from diesel engine should be reduced to keep the clean air environment. PM emission was considered that it consisted of coarse and aggregate particles, and nuclei-mode particles of which diameter was less than 50 nm. However the detail characteristics about these particles of the PM were still unknown and they were needed for more physically accurate measurement and more effective reduction of exhaust PM emission. In this study, the size distributions of solid particles in PM emission were reported. PMs in the tail-pipe emission were sampled from three type diesel engines. Sampled PM was chemically treated to separate the solid carbon fraction from other fractions such as soluble organic fraction (SOF). The electron microscopic and optical-manual size measurement procedures were used to determine the size distribution of primary particles those were formed through coagulation process from nuclei-mode particles and consisted in aggregate particles. The centrifugal sedimentation method was applied to measure the Stokes diameter of dry-soot. Aerodynamic diameters of nano and aggregate particles were measured with scanning mobility particle sizer (SMPS). The peak aggregate diameters detected by SMPS were fallen in the same size regime as the Stokes diameter of dry-soot. Both of primary and Stokes diameters of dry-soot decreased with increases of engine speed and excess air ratio. Also, the effects of fuel properties and engine types on primary and aggregate particle
diameters were discussed.

Key Words: Diesel Engine, Internal Combustion Engine, Fuel, Diesel Exhaust Emission, Particulate, Primary Size, Aggregated Size

1.Introduction

The PM problem in diesel emission is becoming se- rious in the future emission regulations for diesel engines. Especially nuclei-mode particulate matter so called nano- PM is considered that it should be reduced to keep the healthful air environment. Because the health effect(1) – (4) of nano-PM is more serious than the particulate matter of larger size even if the mass contribution of nano-PM is smaller than the larger size PM. The PM measurement including nano-PM was usu- ally performed by the differential mobility analyzer (DMA) + condensation particle counter (CPC) (DMA + CPC = SMPS)(5) – (8), in which a particle size was de-termined as an aero-dynamic particulate diameter. Also there were many other equipments in which different mea- suring principles(9) – (17) were developed corresponding to different properties of particles. PM emission was con- sidered that it consisted of coarse and aggregate carbon

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particles those were enveloped with semi-volatile hydro- carbons, and nuclei-mode particles those were different from primary soot particles consisting in aggregate parti-cles. However, the detail characteristics about the primary and aggregate diameters of the PM are still unknown and they are needed for more physically accurate measurement and more effective improvement of combustion and after-treatment process in diesel engine vehicle.

In this study, the fundamental physical characteris- tics, which could be clarified from electron microscopic

Table 1 Test engines

	E1-DI	E2-IDI	E3-DI
Engine name	Robin DY41D	Ishikawajima-sibaura Test engine	YANMAR NF-D170
Engine type	DI-Diesel 4-stroke, Single Cylinder, OHV, 2-valve	IDI-Diesel 4-stroke, Twin Cylinder, OHV, 2-valve	DI-Diesel 4-stroke, Single Cylinder, OHV, 2-valve
Bore×Stroke[mm]	82×78	90×100	102×105
Displacement[cc]	412	1272	857
Top clearance[mm]	0.9	2	0.85
Cavity volume[cc]	16		38
Comb. Chamber Diameter[mm]	-	34	-
Comb. Chamber Volume[cc]	-	23.1	-
Compression ratio	21	15.8	20.1
Intake valve open	10° BTDC	14° BTDC	
Intake valve close	140° BTDC	138° BTDC	
Exhaust valve open	135° ATDC	128° ATDC	
Exhaust valve close	12° ATDC	10° ATDC	
Valve overlap	22°	24°	
Cooling type	Air cooled	Water cooled	Water cooled

2.Experimental Setup and PM Measurement

Three types of small diesel engines listed in Table 1 were used here. They were air cooled direct injection (DI) diesel engine (E1), water cooled indirect injection (IDI) engine (E2) and water cooled DI diesel engine (E3). A diesel fuel (Light Oil No.2 in Japanese Standard) that had sulfur content of less than 500 ppm was used for E1-DI engine test and low sulfur fuel of less than 50 ppm sulfur was used for E2-IDI and E3-DI engines tests. The exper- imental setup and outline of the PM measurement system are shown in Fig. 1. A conventional dynamometer system was used here. No dilution tunnel was used because of simple labo.-scale test. The PM exhaust emission was di- rectly sampled from the tail pipe of a test engine. It was captured on a quartz-fiber filter (HORIBA QA-100) and analyzed by PM analyzer(12) (HORIBA MEXA-1370) and treated with chemical procedure for extraction of dry-soot. For comparative study of PM size distribution, the exhaust gas contained PM was sampled with a sampling and dilu- tion system (PALAS VKL-100, dilution ratio = 100) and it was analyzed using SMPS (TSI, Model-3034).

The outline of the measurement procedures is de- scribed in Fig. 2. The sampled PM on the filter was collected for the chemical treatment. It was dried-up (110 deg.C, 16 hours) and solved in chloroform solvent to disperse and to extract the dry-soot fraction. To make a specimen for electron microscope, chloroform solvent with PM was dripped on a mesh holder.

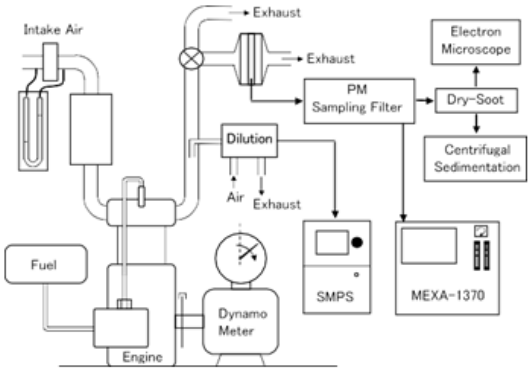

Fig. 1 Experimental setup and PM

photograph were measured by an optical-manual proce- dure using special made measuring system (NIKON Co., Ltd.). For the measurement of cluster size of PM, dried- up PM was again solved in 20%-ethanol water solvent. The Stokes diameter was measured by a centrifugal sedi-mentation method (BROOKHAVEN BI-DCP(type-2)). In this report, aggregate

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diameter of dry-soot was represented with the Sauter mean of Stokes diameter. Sauter mean di- ameter was defined by the volume to surface area ratio of the particles. This mean diameter was usually used for the evaluation of particle behavior related to the surface phe- nomena. In this study, we mainly used the Sauter mean diameter because it seemed that the growth behavior and aggregation of soot particles were related to the surface phenomena.

Since the SMPS measured an aerodynamic diameter of wet-soot and the sedimentation method used here mea-sured a Stokes diameter of dry-soot, it was considered that measured diameters were different with each other. The difference could be explained using a scheme shown in Fig. 3. In a conventional DMA + CPC system like TSI Model-3034, larger particles than 500 nm were eliminated

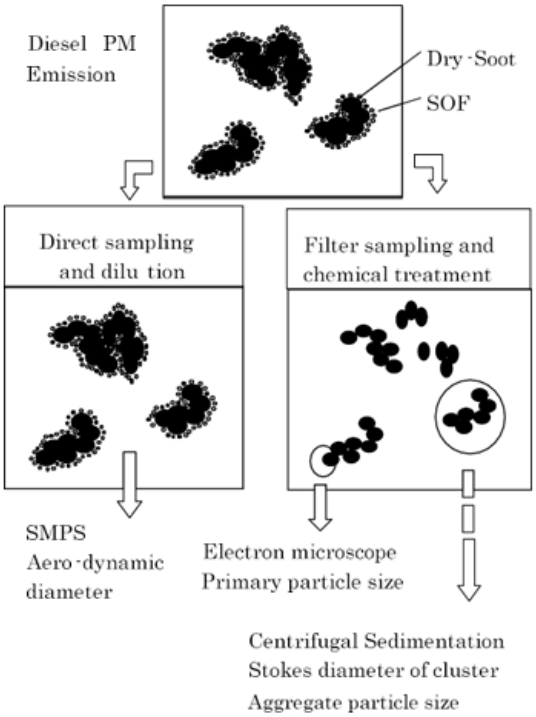

Fig. 2 PM measurement scheme

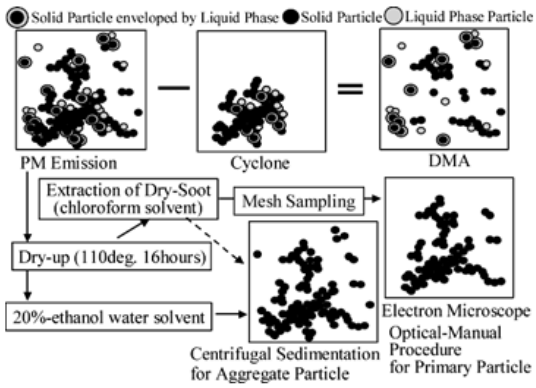

Fig. 3 Primary and aggregate particles in diesel PM emission

3.Primary Particle Size in PM Cluster

A PM cluster usually consisted of many primary par- ticles. Figure 4 is an example of Electron Microscope Pho- tograph of diesel dry-soot. It was captured by the proce- dure mentioned above. In this report, physical meanings of primary and aggregate sizes of PM are defined as shown in Fig. 2. There were clusters consisted of primary parti- cles. Network structure of the clusters had a size larger than 500 nm. It was corresponding to a coarse particle in the PM. However no independent particles smaller than 10 nm could not be observed because of mesh sampling.Figure 5 shows the results of primary particle dis- tributions obtained from the PM emissions from E3 test engine. The number distribution of primary particle size was normalized using total particle numbers and a mea- suring increment of diameter. The operating condition of

λ means the air excess ratio of the engine. Then the λ = 2 means the medium load condition and λ = 1 means the maximum. Low excess air conditions such as λ = 0.7 and λ = 0.5 were surveyed for fundamental study of fuel rich combustion. The primary particle size of PM was distributed from 10 nm to 60 nm. Nano-particles that had the diameter of less than 50 nm was well

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known as nuclei-mode par- ticle(18) and the measured particles were fallen into this size regime. However a particle of its diameter being less than 10 nm was not observed in a microscopic pho- tograph. This is because that such a small particle was too unstable to make a clear image on the microscopic photograph. Further, since the primary particles measured here were coagulated particles those consisted in particle cluster such as shown in Fig. 4, and primary particle was formed from nano-size nuclei-mode particle through a co- agulation process, it was considered that particle clusters could not be directly formed with unstable nuclei-mode particles including liquid phase particles. It had to confirm that dry-soot cluster consisted of the coagulated primary particles shown here. However there was some possibility that large number of nano-size particles were contained in the original PM emissions. As shown in Fig. 5, the excess air ratio had some in- fluence on the size distribution. Lower the excess air ratio, the larger the size distribution shifted to. Especially excess air ratio of λ = 0.5 produced the large primary par- ticles and the mode diameter of it was around 40 nm. Fur- ther, the primary particles at λ = 0.5 had different distri- bution pattern from other three conditions. It was consid-ered that primary particle under extremely fuel rich condi- tion such as λ = 0.5 was formed through different mechan- ism from PM formation mechanism in conventional diesel spray combustion.

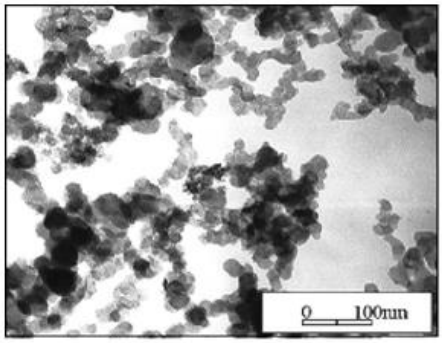

Fig. 4 Electric Microscope Photograph of Dry-Soot E3 Ne = 1080rpm λ = 1.0

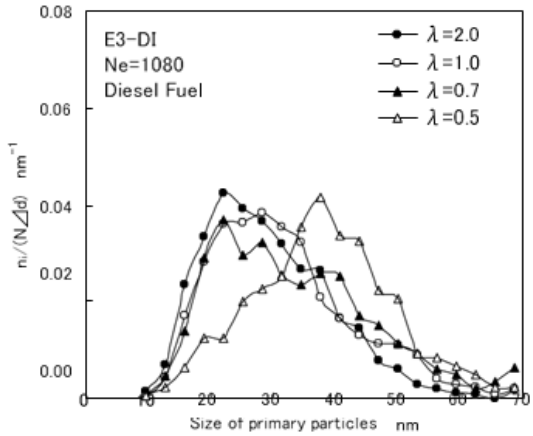

Fig. 5 Primary particle size distributions in aggregate PM cluster

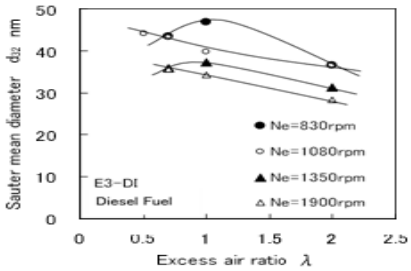

Fig. 6 Sauter mean diameter of primary PM

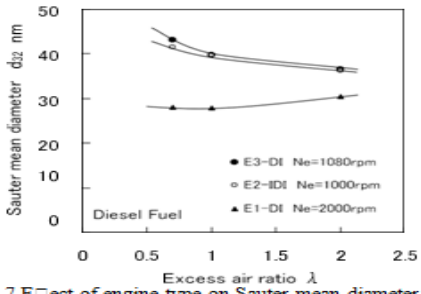

Fig. 7 Effect of engine type on Sauter mean diameter of primary PM

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4.Aggregate Size of PM Cluster

An aggregate diameter of the PM measured by cen- trifugal sedimentation method was considered the Stokes diameter of a cluster that consisted of a large number of primary particles. The Sauter mean diameters D32 of ag- gregate cluster distribution were shown in Fig. 8. The ef- fects of excess air ratio and engine speed on the Sauter mean diameter of aggregate cluster were not so strongly observed at Ne = 830 rpm and 1 350 rpm. However these effects were strongly observed at Ne = 1 080 rpm and 1 900 rpm. The same effect of excess air ratio on aggregate diameter had been reported in the literature(18) and it was considered that the growth rates of primary and aggregate particles were promoted in the fuel rich conditions.From the results of Figs. 6 and 8, it was concluded that the both of primary and aggregate diameters of parti- cles became small with increasing the engine speed and with increasing the excess air ratio.

5. Aerodynamic Diameter Measured by Differential Mobility Analyzer

The measured result of the Stokes diameter shown in Fig. 8 was fallen in a range of accumulation mode di- ameter of the PM emission reported in the literatre(19). Even if the some deference between wet-soot and dry- soot, the Stokes diameter measured by centrifugal sedi- mentation method was considered to be physically equiv- alent with the aerodynamic diameter measured by SMPS. Figures 9–11 show examples of aerodynamic diameter of the PM emission measured by SMPS. Bi-modal distribu- tions were observed for fuel rich conditions and these were different from fuel lean condition (Fig. 11). It was another evidence of different formation mechanism corresponding excess air ratio.

According to the measurement range of SMPS (TSI- Model-3034), it was clear that the larger particles than 500 nm were contained in the PM emissions but it could not be detected. Then, largest peaks in the three conditions were over 500 nm (Figs. 9 and 10) and around 330 nm in Fig. 11. And according the Sauter mean diameter obtained by centrifugal sedimentation method, these larger particles were not affected to the Sauter mean diameters (100 nm– 150 nm). It was concluded that the measurement system of SMPS could detect coarse particles even if it was un- steady and had week network structure that might be de- stroyed in the pre-treatment of centrifugal sedimentation measurement.

The middle peaks of 104 nm in Fig. 9, 111 nm in Fig. 10 and 128 nm in Fig. 11 were almost same as the Sauter mean diameters (Stokes diameters) shown in Fig. 8. This peak diameter was increasing with an increase of ex- cess air ratio.

6.Dry-Soot and SOF Component

The mass of PM exhausted from E3 engine was mea- sured using the PM analyzer (MEXA-1370). The results were summarized in Figs. 12–14. The total PM summa- rized in Fig. 12 shows the dependence of PM emission on excess air ratio. Also the dry-soot in Fig. 13 and SOF in Fig. 14 show the same dependence on

excess air ratio and engine speed. These results were reasonable for conven- tional DI diesel engine. However it is quite interesting that there are no clear correlation between mass of PM and PM size distributions shown in Figs. 6 and 8.

It suggested that the nucleation coagulation and ag- gregation mechanisms were independent from the exhaust mass concentration of PM. From the results of SOF emis- sion, it is easily considered that the SOF component in PM emission plays the important role to stabilize the PM larger than aggregate cluster size. The existence of large coarse particles that were estimated from Figs. 9 and 10, were confirmed with the large SOF fractions shown in Fig. 14.

8.Conclusions

Using the various measurement methods for PM par- ticle emissions, the characteristics of particulate emissions from diesel engine were clarified. The main results were as follows.

(1) Size of primary particle that was coagulated from nuclei-mode particle could be measured by the electron microscope photography method. Stokes diameter of dry- soot could be measured by the centrifugal sedimentation method.

(2) Size distribution of aerodynamic diameter mea- sured with SMPS showed that particles much larger than Stokes diameter consisted in PM emission. But the peaks of aggregate diameters measured with SMPS were fallen in the same size regime as the Stokes diameters sometimes showed the inverse tendency mentioned above.

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