

Particulate Formation in Compression-Ignited Fuel-Rich Mixtures

T.Ogawa, Y.Daisho, O.Kurata* and T.Saito

Department of Mechanical Engineering

Waseda University

4-1 Okubo 3-chome

Shinjuku-ku, Tokyo 169

Japan

** Mechanical Engineering Laboratory*

Agency of Industrial Science and Technology

ABSTRACT

An experimental study has been carried out to investigate the particulate formation in fuel-rich mixtures in the diesel engine. In this work, to avoid complications associated with atomization, heating, evaporation and air-mixing of diesel fuel spray, a single cylinder compression ignition engine system has been developed to ignite and burn homogeneous fuel-rich mixtures which are prepared in the inlet system. In this combustion system, rapid combustion can be reproduced at high pressures and temperatures as observed in diesel combustion.

Pure n-heptane, its blends with cetane and benzene, n-dodecane, and diesel fuel were used for combustion tests. The results show that particulates begin to form when the equivalence ratio reaches about 2.2, and increase greatly with an increase in the ratio. Benzene tends to yield much higher particulate emissions compared to those observed with pure n-heptane. Diesel fuel yields a higher soluble organic fraction (SOF) due to unburned heavy hydrocarbons contained in the fuel. In-cylinder burned gases were extracted by means of high-speed gas sampling and have been analyzed. The results indicate that particulates are formed as the mixture releases heat and then remains constant during the expansion stroke.

INTRODUCTION

Particulates consisting of soot and SOF are among the most undesirable pollutants emitted from the diesel engine. However, little detail is known about the mechanism of particulate formation in the diesel engine due to the highly transient and complicated combustion process taking place at such high pressures and temperatures. Generally, in diesel engines, the spray of fuel injected into the cylinder is heated by the surrounding hot air. It is then evaporated and mixed with the hot air forming heterogeneous mixtures. Because particulate formation and oxidation take place almost simultaneously during the combustion of such mixtures, distinguishing between these two processes is difficult. Numerous investigations⁽¹⁾ have been made to establish soot formation fundamentals using open flames and continuous flow reactors.

In this work, a combustion system has been developed for the specific purpose of reproducing the pressures and temperatures observed in diesel combustion, which are much higher than those used in open flames and reactors. This system uses homogeneous premixed mixtures to avoid the fuel-air

mixing process which inevitably involves the oxidation of particulates. Thus, the effects of equivalence ratios and fuel types on particulate formation were investigated.

EXPERIMENTAL APPARATUS

The combustion system developed includes an inlet system, a single cylinder compression-ignition engine, and measuring instruments as shown in Fig. 1. In this work, homogeneous fuel-rich mixtures were ignited by compression in the engine to avoid complications associated with atomization, heating, evaporation, and air-mixing of the injected fuel spray and to reproduce local fuel-rich regions where diesel particulates were formed. The engine has a flat piston with a 104 mm bore, a 118 mm stroke, and a 7.8 compression ratio. For parametric combustion tests, standard operating conditions were chosen as listed in Table 1. The engine speed and volumetric efficiency were kept constant throughout the tests.

To identify the effects of fuel types, n-heptane was chosen as the simplest reference fuel. This fuel has a cetane number of 56, which yields almost the same compression-ignitability as that of ordinary diesel fuel. N-heptane-benzene and n-heptane-cetane blends, n-dodecane, and JIS No. 2 diesel fuel were also used for comparison. The fuel was metered and injected into the inlet pipe by means of an electronically controlled fuel injection system. The injected fuel was heated and evaporated by preheated air and then fully mixed with the air in the mixing chamber.

The inlet temperature of the fuel-air mixture was adjusted to within the range of 160 to 200°C by changing the input of the electric heater. Such high inlet temperatures allowed the mixture to be ignited by compression even at the low compression ratio. Thus, stable and repeatable compression-ignition and combustion were achieved by using fully premixed homogeneous mixtures at high temperatures⁽²⁾.

A piezoelectric pressure transducer was used to determine the ignition timing, the mean temperature, and the rate of heat release. Gas analysis was

Table 1 Standard operating conditions

Engine speed	1,000 rpm
Volumetric efficiency	50 %
Inlet temperature	170 °C
Fuel	N-heptane
Equivalence ratio	2.9

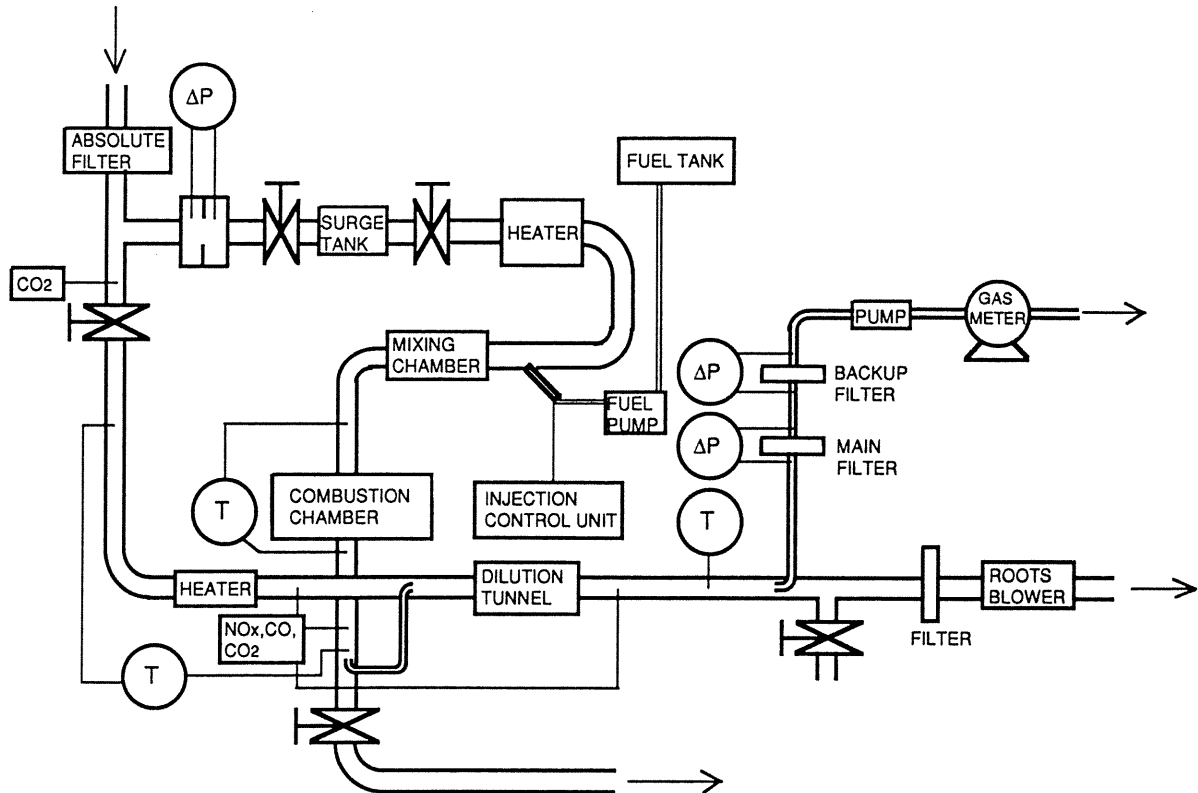


Fig. 1 Schematic of the experimental apparatus

performed for sampled exhaust gases by gas chromatography. A mini-dilution tunnel with filters was employed to sample and to weigh the particulates. SOF and solid soot were then separated using dichloromethane as a solvent.

Further, in order to examine the combustion process and the particulate formation, in-cylinder gases were extracted using a high speed gas sampling system having two electronically actuated needle valves⁽³⁾. One was actuated just prior to the opening of the other one in order to remove any gas remaining in the sampling line. Samples were instantaneously diluted with helium and then analyzed to determine the compositions and the particulate mass.

RESULTS AND DISCUSSION

Effects of Equivalence Ratio

N-heptane, n-dodecane, and JIS No. 2 diesel fuel were burned in the combustion system at different equivalence ratios, and exhaust particulates were analyzed. As shown in Fig. 2, in n-heptane and n-dodecane mixtures, exhaust soot began to appear at an equivalence ratio of 2.2 and then rapidly increased with increasing ratios. However, SOF emissions were low for all the equivalence ratios tested. By contrast, in the case of the diesel fuel, the SOF level was high and increased with the equivalence ratio whereas the soot level decreased. This suggested that SOF

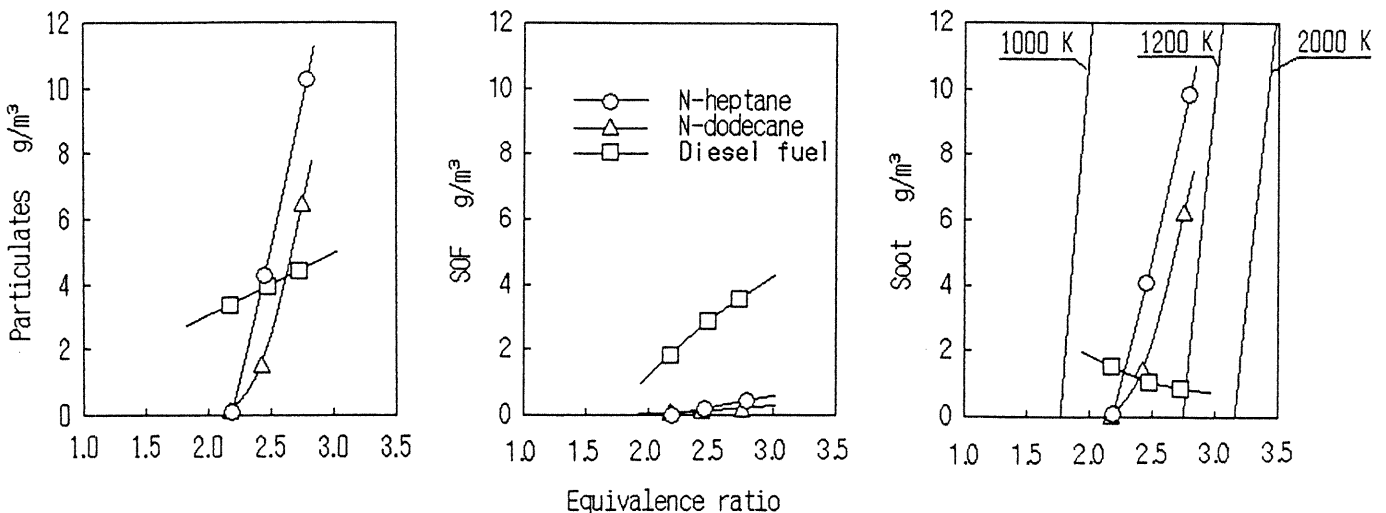


Fig. 2 Effects of equivalence ratio

results from unburned fuel. More detailed discussion on this result will be made later.

Figure 3 shows the cylinder pressure diagrams, mean gas temperatures, and the rates of heat release for n-heptane mixtures at three equivalence ratios. Peak pressures range from 3.0 to 3.3 MPa, peak temperatures from 1,900 to 2,100 K, and combustion periods from 1.0 to 2.0 ms. These conditions are almost equivalent to those in fuel-rich regions where particulates are formed in diesel combustion. Small peaks of heat release rates appear at the early combustion stage. These are caused by lower temperature chemistry inherent in compression-ignition of hydrocarbon mixtures. Since combustion in this system can be regarded to be spatially uniform, the heat release period is equivalent to the chemical reaction time. In combustion in a closed system, reaction heat will raise pressures and temperatures to much higher levels compared to those reached in experiments using open flames and continuous flow reactors. Thus, in this combustion system, rapid combustion can be reproduced at high pressures and temperatures for simulating diesel combustion.

As can be seen from Figs. 2 and 3, soot increases with the equivalence ratio although pressures and temperatures are lowered. This suggests that the equivalence ratio is the most important factor influencing soot formation at the pressures and temperatures reached in this combustion system. Solid carbon densities at the equilibrium state were calculated for n-heptane mixtures at a pressure of 3 MPa and different temperatures, taking into account twenty one

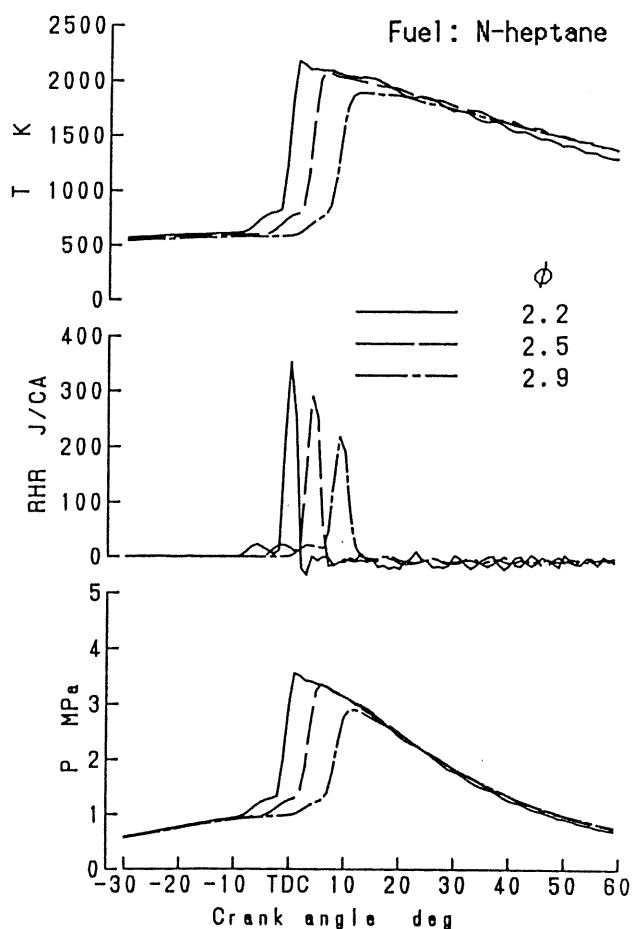


Fig. 3 Cylinder pressures, temperatures and rates of heat release for n-heptane mixtures

chemical species including up to C_2 hydrocarbons. Obviously, real particulates are formed at a lower equivalence ratio than that for equilibrium carbon at 2,000 K. However, in both cases, the dependence of soot formation on the equivalence ratio has some similarity.

Effects of Fuel Types

N-heptane-benzene and n-heptane-cetane blends were then compared at an equivalence ratio of 2.9. In Fig. 4, relative soot and SOF levels are shown as functions of benzene and cetane added by volume. The figure shows that benzene, the simplest aromatic hydrocarbon, has the stronger effect on particulate formation by a factor of three to four compared to n-heptane. Benzene tends to increase relative SOF levels more than it increases the soot level although SOF levels were low. This suggests that aromatic compounds will form soot much faster.

In Fig. 5, relative soot levels are shown as a function of carbon mass fraction in each fuel. Calculated equilibrium carbon levels are also shown in this figure. The carbon mass fraction partly explains the reason for the increase in particulates with blending benzene. On the contrary, as far as the paraffinic hydrocarbons are concerned, heavier fuels will form less soot and SOF despite the increase in carbon mass fraction. This tendency can also be seen in the comparison between n-heptane and n-dodecane in Fig. 2. This implies that for soot formation, heavier hydrocarbons must be fragmented

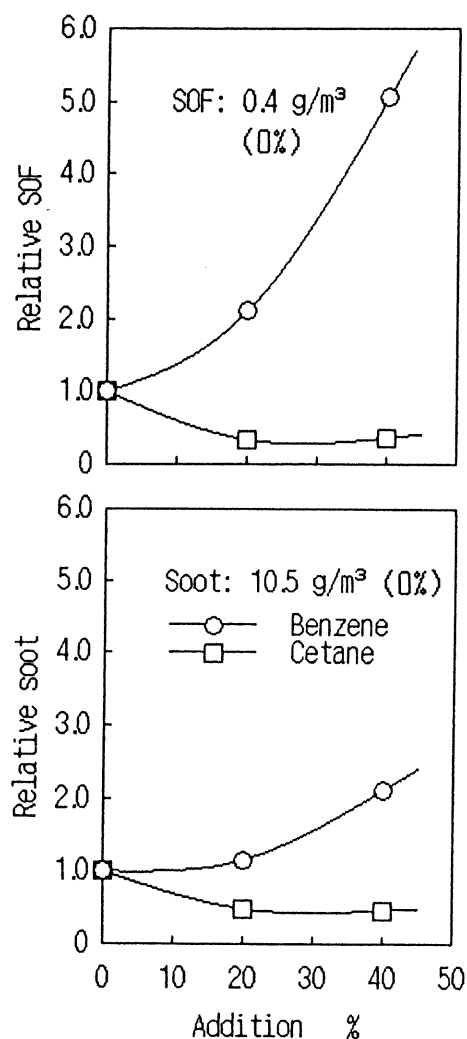


Fig. 4 Comparison of fuels added to n-heptane

into lighter hydrocarbons by some chemical course.

Figure 6 shows the cylinder pressures, the mean gas temperatures, and the rates of heat release for the different fuels. As seen from this figure, although benzene tends to delay ignition by a few crank angle degrees, it does not significantly change the rate of heat release, the peak pressure, or the peak temperature. In cetane blends, the amount of particulates is lowered although ignition timing is advanced and pressures and temperatures are increased. Thus, soot formation chemistry seems to be quite different depending on whether or not the fuel contains aromatics.

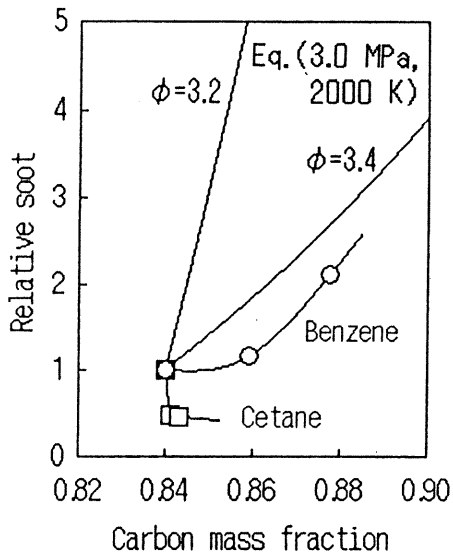
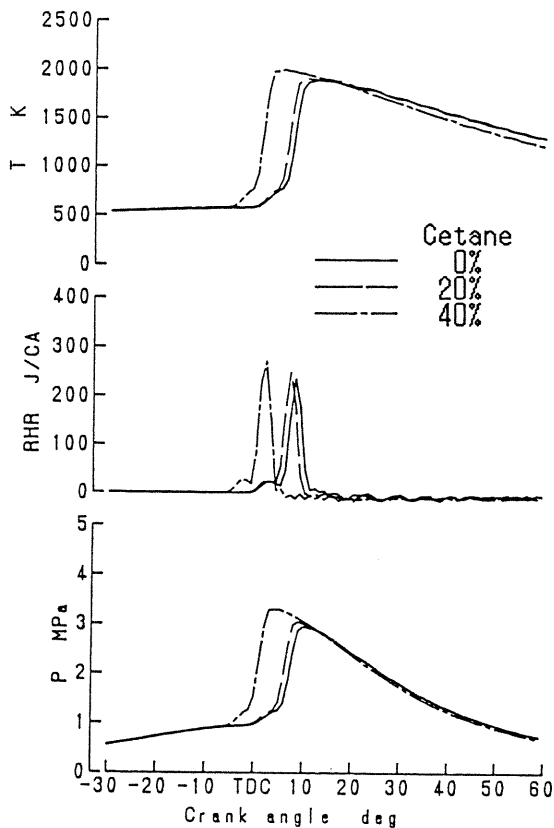


Fig. 5 Effects of carbon mass fraction in fuels



In-Cylinder Gas Sampling and Analysis

High-speed, in-cylinder gas sampling was performed to analyze particulates and other species in the combustion of n-heptane and diesel fuel. Figure 7 shows the plots of soot and SOF analyzed for different crank angles. Each graph is plotted for the middle of the 1.0 ms valve opening duration. As can be seen from this figure, in n-heptane combustion, soot formation occurs as fast as the rate of heat release and then remains constant during the expansion stroke. This fact confirms that the combustion system using fuel-rich mixtures can reproduce the soot formation process without oxidation.

In the case of diesel fuel, high SOF levels, which are due to the heavy part of the fuel, are observed prior to combustion. Part of the fuel remains unburned after combustion and is emitted in the exhaust gas. Table 2 shows the composition of the diesel fuel used in this work. Some amount of soot and SOF emissions may result from these aromatics.

Figure 8 shows the results of light hydrocarbon analysis for the two fuels. As the source of soot, unsaturated hydrocarbons such as acetylene, ethylene, and propylene, which are formed due to the fragmentation of the fuel, are observed to increase with the progress in heat release. Higher concentrations of such species are obvious in n-heptane.

Hydrocarbon compositions were then analyzed for the diesel fuel and its exhaust SOF using high temperature gas chromatography. Figure 9 indicates that SOF emissions come from heavy hydrocarbons which in spite of being exposed to high temperatures are not burned due to the very short residence time and lack of oxygen in the combustion system. By contrast, in n-heptane combustion, such heavy hydrocarbons were not detected.

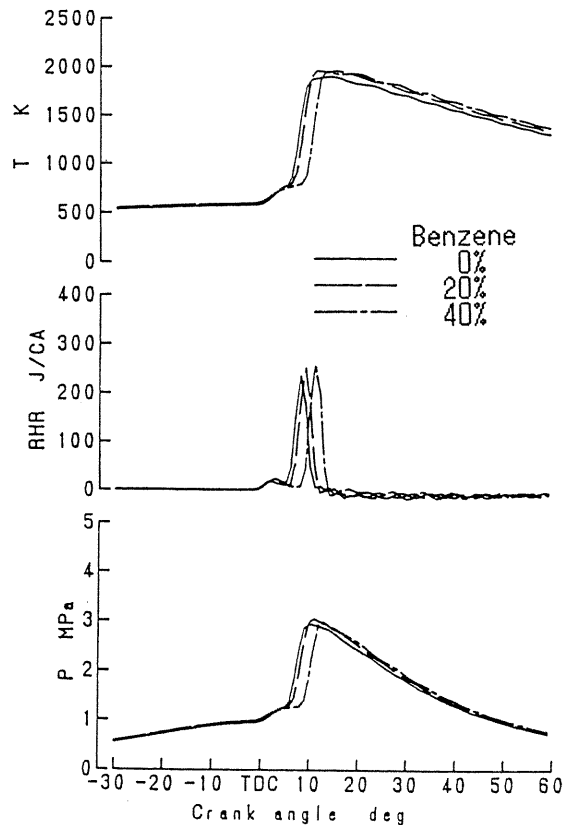


Fig. 6 Cylinder pressures, temperatures and rates of heat release for different fuels

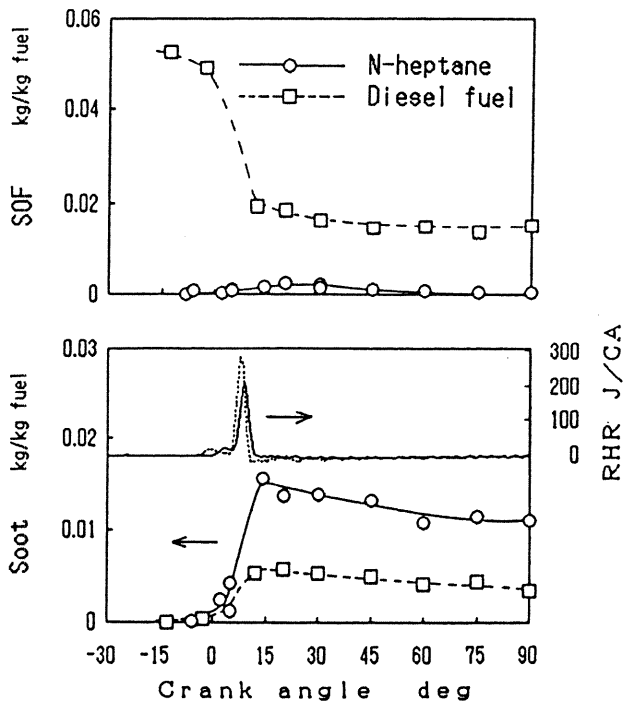


Fig. 7 In-cylinder soot and SOF

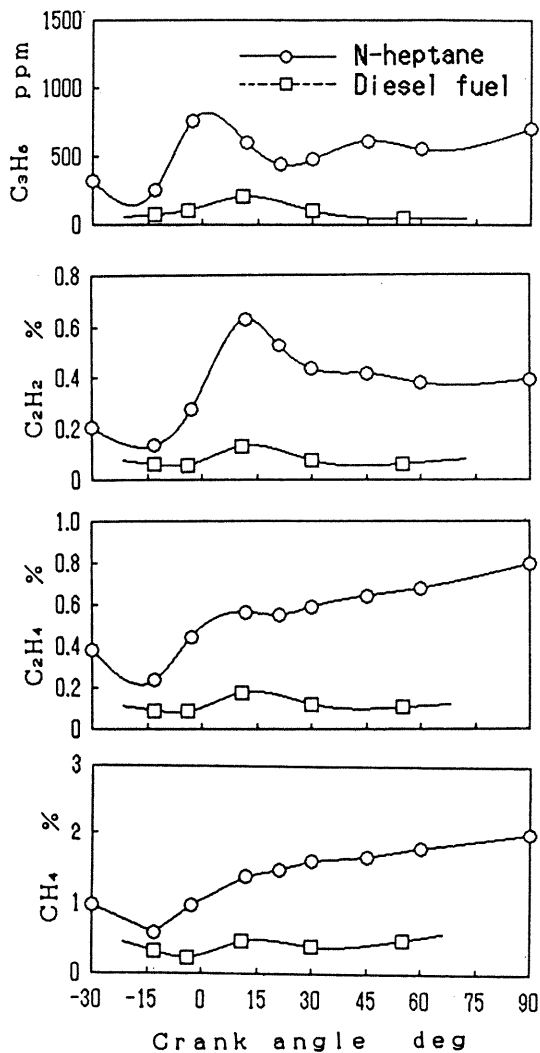


Fig. 8 In-cylinder light hydrocarbons

Table 2 Composition of the diesel fuel used

C: 85.6	H: 13.7	N: <1.0	S: 0.43 % (wt)
Saturated hydrocarbons: 69.4 % (wt)			
One-ring aromatics: 21.3			
Two-ring aromatics: 9.1			
Other aromatics: 0.2			

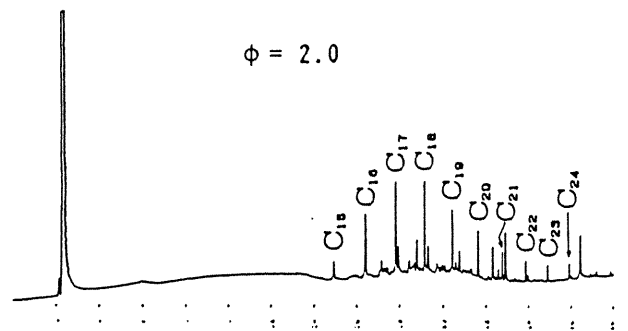
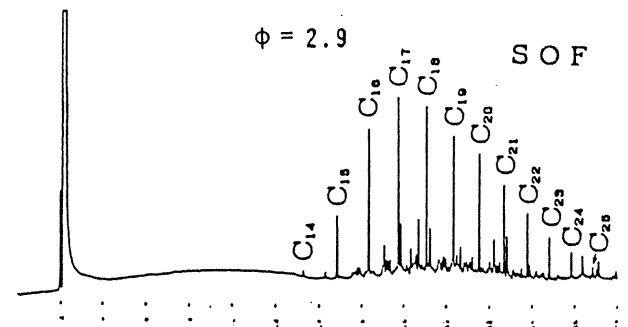
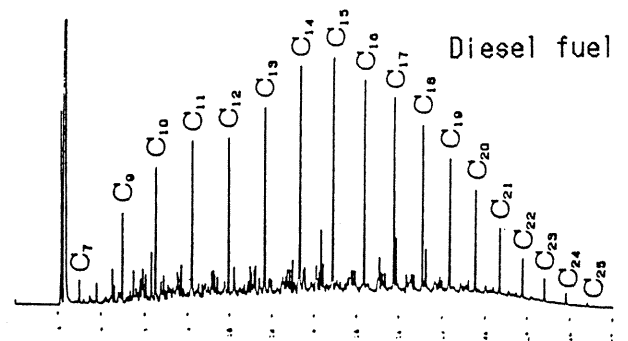


Fig. 9 Hydrocarbon compositions in diesel fuel and SOF

Because the soot formed in n-heptane is associated with heat release and then remains constant as shown in Fig. 7, an activation energy is roughly estimated for the overall soot formation reaction assuming that the constant soot levels are governed by peak temperatures. Peak temperatures and soot levels were changed, by adjusting the inlet temperature and keeping other operating parameters constant. Figure 10 shows the Arrhenius plots. From this result, the activation energy are estimated to be 240 kJ/mol.

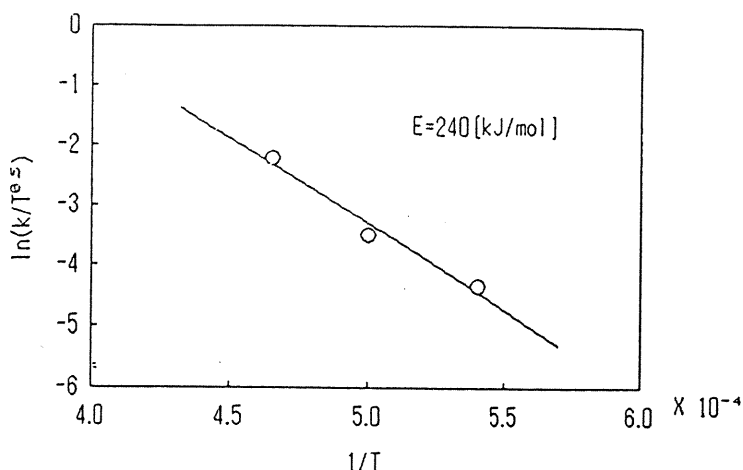


Fig. 10 Arrhenius plots for soot formation

CONCLUDING REMARKS

In order to reproduce the particulate formation occurring in fuel-rich mixtures in diesel combustion, a combustion system has been developed using a single cylinder compression-ignition engine fueled with homogeneous premixed mixtures. In this combustion system, rapid combustion can be reproduced at pressures up to 3.5 MPa and temperatures up to 2,100 K as observed in diesel combustion.

The combustion test results indicate that the equivalence ratio and the aromatics content in the fuel are dominant factors influencing the particulate formation at the pressures and the temperatures tested. The heavier paraffin hydrocarbons tend to form less soot than the lighter ones. In diesel fuel combustion, SOF emissions result mainly from some heavier part of the unburned fuel.

In combustion of paraffin hydrocarbons, SOF levels are much lower than soot levels. In-cylinder gas analysis shows that particulate formation is associated with heat release of the fuel and then during constant due to the lack of oxygen. Based on this fact, an activation energy of 240 kJ/mol for the overall sooting reaction in n-heptane mixtures has been estimated.

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