

A Photographic Study of Soot Formation and Combustion in a Diesel Flame with a Rapid Compression Machine

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ABSTRACT

The formation and oxidation processes of soot particles in a diesel flame were investigated with a rapid compression machine. A cloud of soot particles was successfully visualized by means of the instantaneous laser schlieren technique and the equivalence ratio of the soot formation zone was estimated from a measured fuel concentration distribution in a non-evaporating spray. The temporal and spatial variation of soot concentration in the flame was also correlated with the rate of heat release.

Soot particles appear first in a region near the flame tip when diffusion combustion period starts, and its concentration is a maximum at about the end of injection, then decreases due to oxidation. The reason for soot being formed in a fuel lean region near the flame tip is that the evaporated fuel requires time to be pyrolyzed as it travels through the burning fuel rich zone towards the flame tip.

INTRODUCTION

Soot emission seems to be an inevitable and critical phenomenon in diesel engines which employ the diffusion type combustion. Hence, there is no doubt that the reduction of soot emission will remain to be one of the principal subjects in the continuing combustion research in diesel engines.

Formation and oxidation processes of soot particles in the combustion chamber of a DI diesel engine were investigated by means of both the direct gas-sampling method by Aoyagi et al.(1) and the two-color method by Matsui et al.(2). The time histories of soot concentration measured by both methods revealed that soot formation starts simultaneously with the onset of diffusion combustion, and that its concentration increases rapidly with time during the injection period. This concentration reaches a maximum at about the end of injection. Soot is then oxidized at a rapid rate down to a concentration of about one fiftieth of the maximum concentration, but after the flame temperature drops below 1800 K the oxidation nearby stops and the soot is then exhausted at that concentration level. Using their experimental data, Kamimoto et al.(3) calculated the soot oxidation rate in the diesel flame and found that it is rate-controlled by the oxygen partial pressure around the soot particles, namely due to turbulent mixing.

From these experimental observations the formation and oxidation processes in diesel flames were made clear to a much greater extent, however the key formation process whereby injected fuel is converted into soot particles in the flame remains to be solved because the flame in the combustion chamber moves in a complicated manner in the small confined space being affected both by the walls and air motion.

In addition to the above experimental studies, prediction of soot emission from diesel engines has already been pursued by many researchers. Nevertheless, no suitable models for soot formation as yet exists, so that different assumptions have been made by each researcher. Khan et al.(4) assumed that soot is formed only in the fuel rich core of spray according to an Arrhenius type of reaction rate thereby neglecting the soot oxidation process. Ikegami and Shioji(5) who developed a stochastic mixing model for the prediction of heat release in a DI diesel engine, assumed that the chemical equilibrium calculation can be applied quasi-steadily for the prediction of soot concentration of local mixture. The equivalence ratio of each mixture element, i.e. the non-homogeneity of mixture, was estimated by means of their stochastic model. Mansouri et al.(6) predicted soot emission from an IDI diesel engine with their own stochastic mixing model. The initial soot mass loading which was determined from the chemical equilibrium calculation, decreases by oxidation at a rate which is estimated by the Nagle and Strickland-Constable formula(7). Hiroyasu et al.(8) also calculated soot concentration in the flame by employing their own mathematical model for an IDI diesel engine. They used an Arrhenius type of equation for soot formation rate basing upon their own experiments and estimated an oxidation rate by the Nagle and Strickland-Constable formula.

Both the chemical equilibrium and an Arrhenius type kinetic assumptions on which these existing models are based state that the more soot is formed, the greater the mixture strength is increased. However, this concept seems to be in conflict occasionally with the observations of high speed photographs of a diesel flame. Actually radiant luminosity, which is caused by soot particles in the flame is very weak at a region near the nozzle orifice where equivalence ratio is at its highest.

This paper presents these objectives; to interpret both the soot concentration data obtained by the gas sampling technique, and the flame luminosity

observed on the high speed photographs, specifically to understand the formation and oxidation processes of soot in the diesel flame; and to assess the assumptions on soot formation appearing in each simulation models described above. The processes about when and where soot is formed and burned out were successfully visualized via the Schlieren technique with a diesel flame in a quiescent atmosphere in a large rapid compression machine at the Tokyo Institute of Technology. The fuel concentration distribution in a non-evaporating spray was measured by means of an image analysis technique, and the equivalence ratio in the soot yielding region was determined.

MEASUREMENT PRINCIPLE

For the measurement of soot concentration in a diesel flame, two kinds of measuring method were employed. One is a film image analysis method which provides an instantaneous and qualitative information on the whole structure and soot concentration in a flame. Another is a laser beam extinction method which gives a quantitative data on local soot concentration in a flame. Both methods are based on the incident light extinction principle.

When a parallel light with an intensity I_0 is incident to a flame, i.e. a cloud of soot particles, the intensity of the parallel light behind the flame is decreased to I . The emission from the flame is cut by a narrow band pass filter. If particles with the same diameter D are uniformly and thinly dispersed, the degree of attenuation, which is defined as the transmission, is expressed by the following equation (9).

$$\tau = I/I_0 = \exp(-Q_{\text{ext}} \frac{\pi D^2}{4} NL) \quad (1)$$

where N is the number density and L is the thickness of soot cloud and the extinction coefficient of soot particles Q_{ext} is represented as follows in the case of $\pi D/\lambda \gg 1$

$$Q_{\text{ext}} = \frac{24\pi D}{\lambda} f(n,k) \quad (2)$$

where λ is the wavelength of incident light, and $f(n,k)$ is a function of the complex refractive index of soot particles $n + ik$, and it can be written as follows

$$f(n,k) = \frac{nk}{(n^2+k^2)^2 + 4(n^2-k^2) + 1} \quad (3)$$

While the concentration of soot particles, C_S (kg/m³) is given by

$$C_S = \rho_S \frac{\pi}{6} D^3 N \quad (4)$$

where ρ_S is the density of soot particle. From equations (1), (2), and (4), C_S can be calculated from the measured transmission as follows.

$$C_S = \frac{\rho_S}{36\pi L f(n,k)} \log \tau \quad (5)$$

Where $m = 1.9 - 0.35i$, which was proposed by Chipplet and Gray(10) was used, and ρ_S is assumed to be 1.8 g/cc(11).

EXPERIMENTAL APPARATUS AND METHODS

Rapid Compression Machine

Fig. 1 shows the rapid compression machine of Tokyo Institute of Technology(12). It is actuated by high pressure nitrogen of 8 MPa and the piston motion is controlled by a hydraulic system. The combustion chamber of the rapid compression machine is of the pancake type with a diameter of 196 mm and a thickness of 40 mm. The stroke is 560 mm and the compression ratio is 14.7. Photographs of spray and flame were taken through the glass windows accommodated within the combustion chamber. The single shot injection of the fuel was performed by pulling the rack of an injection pump using an air actuator. Two injection directions can be selected as shown in Fig. 2. Free diesel flame, i.e., a diesel flame in a quiescent atmosphere was mainly examined. The effects of impingement on soot formation were also investigated at the mode shown in Fig. 2.

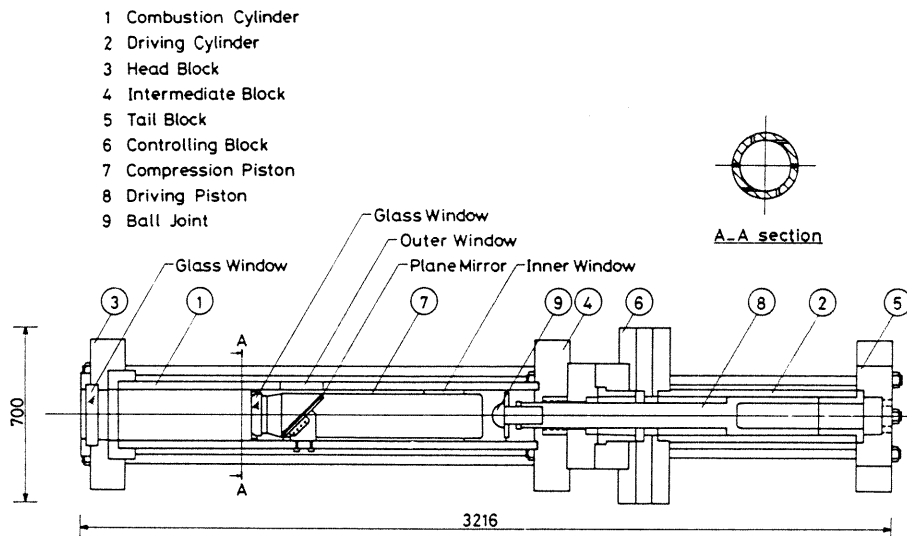


Fig. 1 Rapid compression machine at Tokyo Institute of Technology

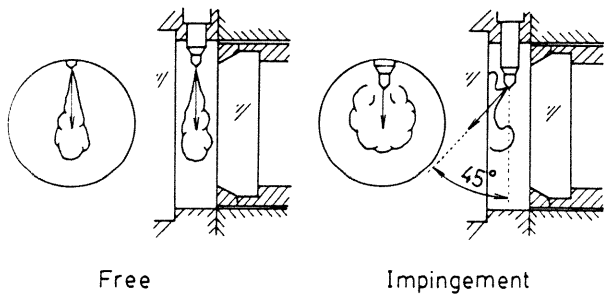


Fig. 2 Direction of fuel injection for free flame and impinging flame

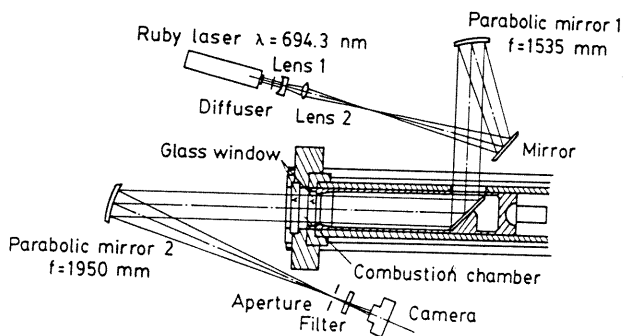


Fig. 3 Optical arrangement of instantaneous schlieren photography using pulse laser

Schlieren Photography

Fig. 3 illustrates the optical arrangement for the schlieren photography. A pulsed ruby laser with a wavelength of 694.3 nm, output power of 40 MW and a pulse duration of 25 ns was used as the light source. Laser beam is expanded by lens 1 and 2 and is made parallel by parabolic mirror 1. Some portion of incident light is attenuated by soot particles in the flame, and the transmitted light is converged by parabolic mirror 2 to form an image of the light source at the focal plane of parabolic mirror 2. An aperture was placed at the focus of parabolic mirror 2 to adjust the schlieren effect due to gas density gradient in the combustion chamber. Between the aperture and camera, a narrow band pass filter, which has a central wavelength of 694.3 nm and a half width of 5 nm, was placed in order to remove the self emission from the flame. Two sheets of mylar film were inserted between the laser and lens 1 to adjust the light intensity and to diffuse the laser beam. A uniform illumination without speckles was achieved with this arrangement. A 35 mm camera with a lens whose focus length is 300 mm was utilized. A panchromatic type (Fuji: Neopan 400) with a sensitivity of ASA 400 was used.

Since the scattered light by soot particles can be ignored as compared with the absorbed light, the aperture dimension is not responsible for the measurement of extinction by soot particles. But the schlieren effect brought about by the gas density gradient in the combustion chamber becomes more significant as the aperture size becomes smaller. In order to pay attention only to the extinction by soot particles, the aperture dimension

should be large enough to neglect the schlieren effect. Since soot formation must be discussed in relation to the behavior of a flame, the observation of the development of the whole flame is therefore required. For these reasons the aperture size should be carefully selected. Fig. 4 shows the effect of aperture size on the obtained photographs. With the largest size of aperture only the shadow of the cloud of soot particles was caught, whereas the smallest size of aperture allowed the visualization of density unevenness. Considering the objectives of this study, the aperture with a diameter of 10 mm was selected.

Laser Beam Extinction Method

For quantitative measurements of soot concentration in a diesel flame, measurements by the laser beam extinction method were conducted using an optical arrangement shown in Fig. 5. A He-Ne laser with a wavelength of 632.8 nm, a power of 25 mW, and a beam diameter of 1.3 mm was utilized. The laser beam is chopped at a frequency of 2 KHz by a mechanical chopper which is located in front of the laser. The attenuated laser beam after passing through a soot cloud in the flame hits a diffuser; mylar sheet, placed at a distance of 90 mm from the spray axis. The diffused light was led through an optical fiber to a photomultiplier. The transmission τ was determined by the absorption - emission relation:

$$\tau = (I_t - I_f) / I_0$$

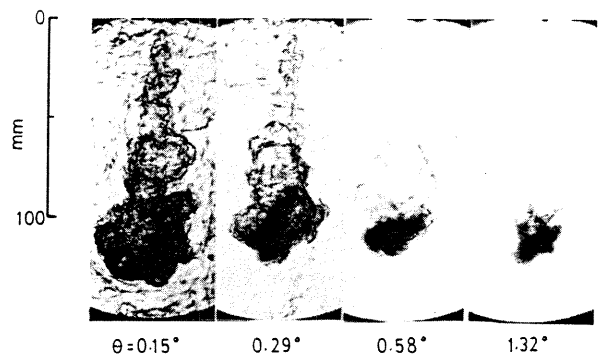


Fig. 4 Effect of aperture size on sensitivity of density gradient in flame

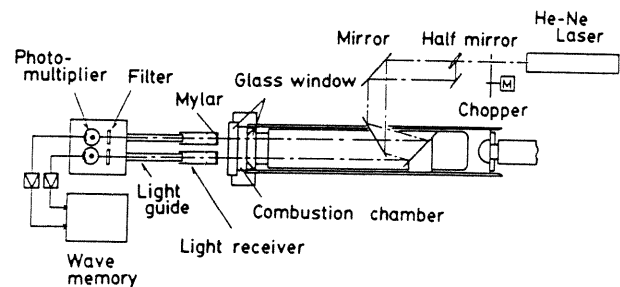


Fig. 5 Optical arrangement of laser extinction method for quantitative measurement of soot concentration in flame

where I_f is the intensity of detected light when the chopper is closed, i.e. the intensity of self emission from the flame, and I_t is the intensity of detected light when the chopper is opened, i.e. the intensity of transmitted laser beam plus I_f .

Experimental Conditions

Table 1 shows the injection condition and Table 2 indicates the surrounding air conditions at the onset of fuel injection. Fig. 6 shows the injection rate corresponding to the conditions listed in Table 1. A spray in a high-pressure nitrogen atmosphere at a room temperature is designated as "non-evaporating spray", and a spray in the high pressure and temperature nitrogen is designated as "evaporating spray". A flame with a short ignition delay, a flame with a long ignition delay, and an impinging

Table 1 Injection conditions

Fuel	n-tridecane
Nozzle opening pressure	21.7 MPa
Nozzle dia.	0.2 mm
Inj. duration	3.0 ms
Fuel amount	16.0 mg
l/d	3.0
Pump speed	750 rpm

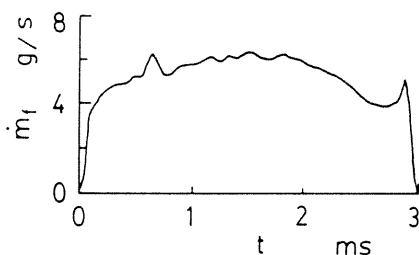


Fig. 6 Rate of fuel injection at condition shown in Table 1

flame as shown in Fig. 2 are named "Flame S", "Flame L", and "Flame I" respectively. Variation of ignition delay with initial temperature was made possible by means of pre-heating the supplied air.

EXPERIMENTAL RESULTS AND DISCUSSIONS

Results

Soot formation zone. Fig. 7 shows a series of instantaneous photographs of spray and flame. Fig. 8 shows direct photographs of Flame S. The rates of heat release for Flame S and Flame L, as presented in Fig. 9 for convenience, in relation with soot formation which will be described later. In these figures, t means the time after the start of fuel injection.

A comparison between the non-evaporating spray and evaporating spray shows that the latter has brighter shadow near the spray tip region, which implies the existence of fuel vapour thereat. In the case of evaporating spray, a dark slender part near the nozzle exit can be observed, which shows the existence of fuel droplets or liquid core, the length of which is around 40 - 50 mm corresponding to the distance required for the injected fuel to break-up and evaporate. A comparison between evaporating spray and flames indicates that during the injection period, the length of a dark shadow near the nozzle is shorter for flames, which is probably due to a high gas temperature in the flame; and that a zone accompanied with a dark shadow is notable near the tip of Flames S and L which cannot be observed in the evaporating spray.

A dark shadow observed near the tip in the case of Flame S corresponds to the luminous zone shown in Fig. 8. From the above observations, the dark shadow near the flame tip is considered to be due to the extinction by soot particles. In order to substantiate this concept, an experiment was conducted for a flame by means of the laser extinction method. The conditions of the fuel injection and surrounding air being subjected to the experiment are as follows: nozzle diameter = 0.14 mm, $l/d = 3.0$, injection duration = 3.05 ms, mean injection rate = 2.46 g/s, mean injection pressure = 22.8 MPa; air pressure = 3.05 MPa, air temperature = 737 K, and air density = 14.4 kg/m³. The direct high speed photographs for the flame are shown in Fig. 10. Fig. 11 shows the measured results of soot concentration distribution C_s along the spray axis with t as a parameter, as well as the time history of soot formation ratio R_s , which is defined as the mass ratio of soot to the carbon in the injected fuel. It turned out from Figures 10 and 11 that high soot concentration zone corresponds generally to the

Table 2 Surrounding atmosphere conditions

Series	Atmosphere	Initial temp. T1 K	Pressure P2 MPa	Temp. T2 K	Density ρ_2 kg/m	Ignition delay ms
Non-evaporating spray	N2	--	1.24	286	15.1	--
Evaporating spray	N2	340	2.93	700	14.7	--
Flame S	Air	320	3.23	720	15.7	1.0
Flame L	Air	300	3.13	660	16.7	1.8
Flame I	Air	320	3.08	690	15.6	1.3

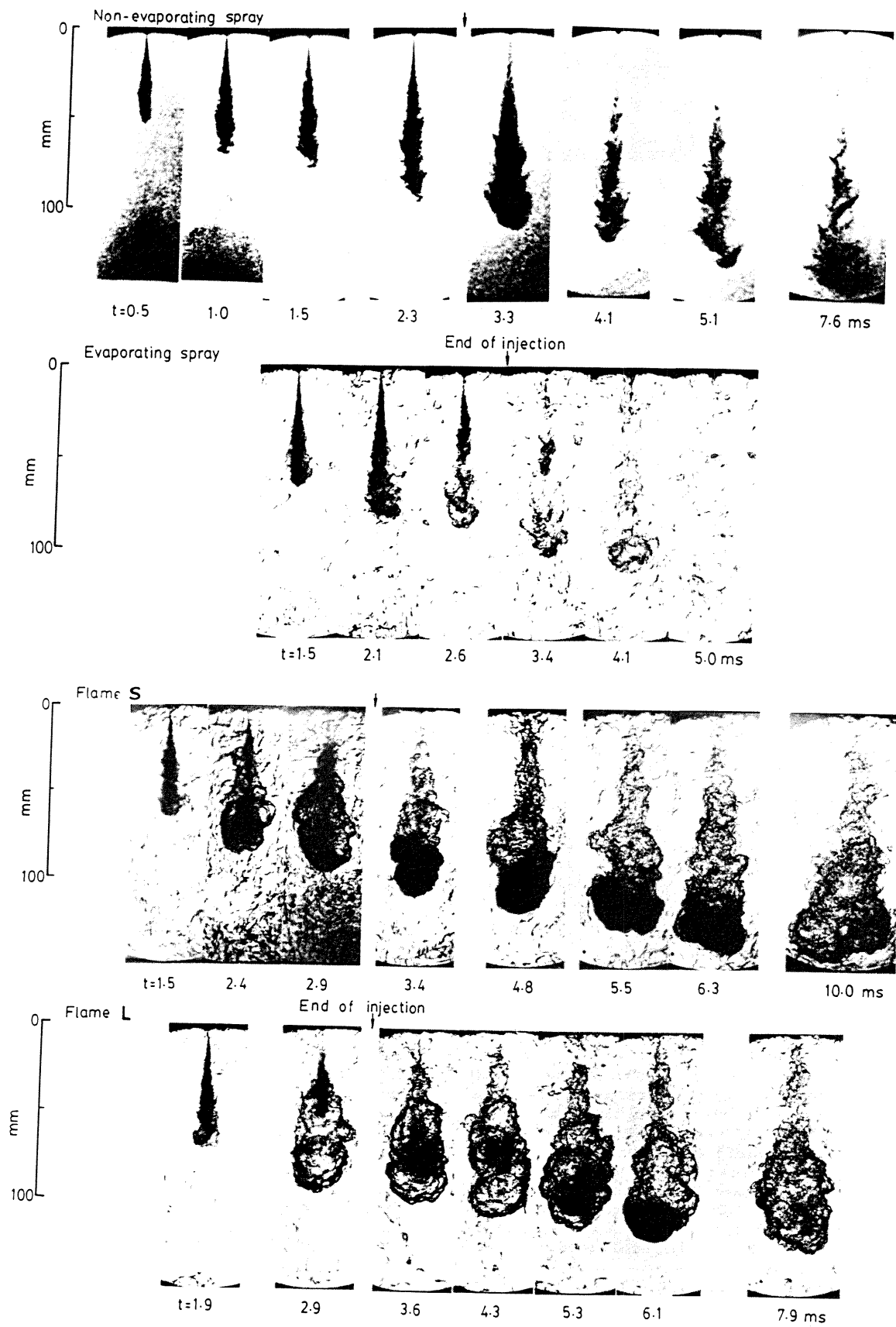


Fig. 7 Series of instantaneous photographs of non-evaporating spray, evaporating spray, Flame S and Flame L

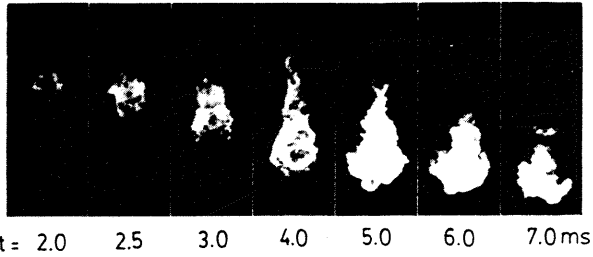


Fig. 8 Direct high speed photographs for Flame S

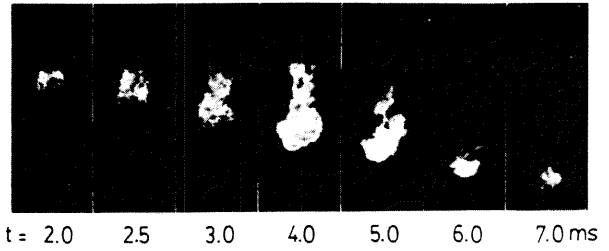


Fig. 10 Direct high speed photographs of a flame (Conditions: see text)

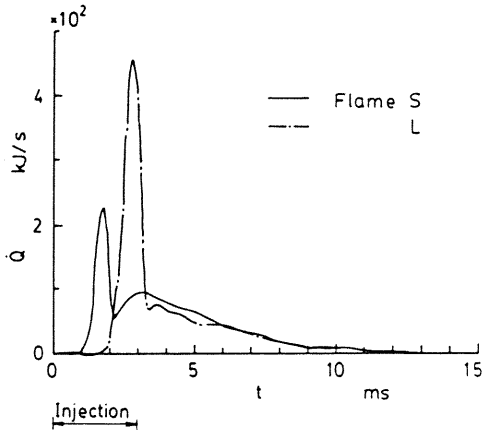


Fig. 9 Rate of heat release for Flame S and Flame L

luminous part of the flame. Thus, we can conclude that the dark shadow observed on the schlieren photographs indicates the existence of soot particles. Basing on this result, it is obvious that soot is formed only in a region which is about 50 mm away from the nozzle orifice and that C_s takes a maximum value at $t = 3 - 4$ ms, which is followed by a gradual decrease due to oxidation of soot particles. Apart from the above discussion it is notable in Fig. 11 that soot formation ratio R_s amounts to as high as 20 percent at 1 ms after the end of injection.

Relation between soot formation and heat release. Comparing Schlieren photographs shown in Fig. 7 and the rate of heat release shown in Fig. 9, we would like to investigate the relation between soot formation and heat release.

In the case of Flame S: During the initial combustion period, no soot was detected in the flame. Soot is observed for the first time immediately after the start of diffusion combustion period and the sooting zone rapidly expands its volume toward the flame tip. When the rate of heat release reaches its peak, the magnitude of sooting zone also reaches its maximum. Thereafter the magnitude of sooting zone remains constant for a while, soot is subsequently oxidized or diffused gradually and finally extinguished.

In the case of Flame L: It is noted from photographs that Flame L, i.e. a flame with a longer ignition delay, yields less soot than Flame S. Longer ignition delay leads to a higher initial heat release because of sufficient time for mixture preparation. Our previous paper(13) revealed that a

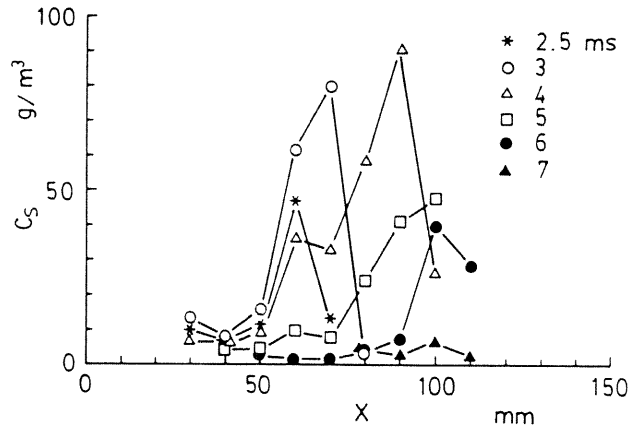
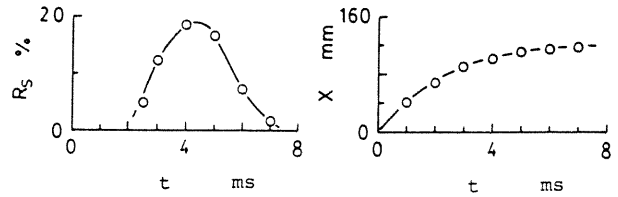


Fig. 11 Variation of soot concentration C_s along spray axis against distance X from nozzle orifice with time as a parameter, variation of conversion rate of carbon to soot R_s with time, and tip penetration versus time (conditions : same as in Fig. 10)

rapid thermal expansion of flame volume occurs during initial combustion period due to a high heat release, and that this rapid expansion promotes mixing and air entrainment. It is probable that higher mixing rate in Flame L suppresses soot formation and promotes soot oxidation.

Impinging flame. Fig. 12 illustrates the schlieren photographs of an impinging flame, Flame I, and its heat release rate. Impinging distance in this case is 31 mm, which is shorter than the break-up and evaporation length of free spray flames shown in Fig. 7. Though the ignition delay in this case lies between Flame S and Flame L, the amount of soot formed appears to be slightly less than Flame L. The mixing process must have been promoted by the wall impingement. Promotion of mixing process in Flame I can be also confirmed by the shorter combustion period and by the higher rate of heat release than that for free spray flames.

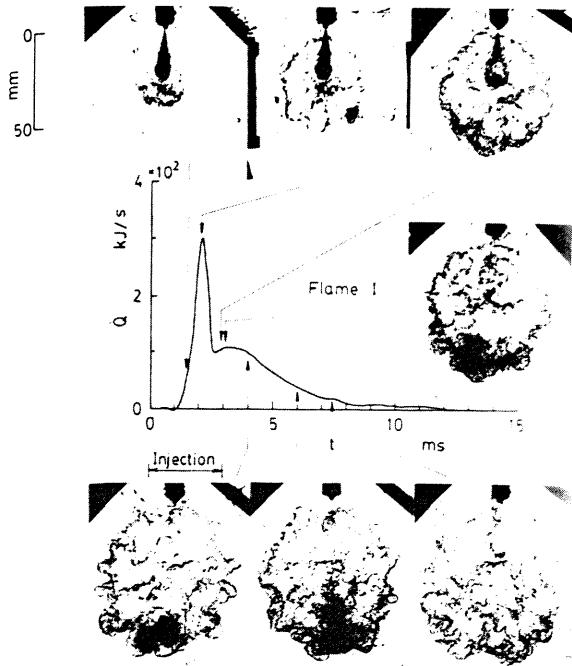


Fig. 12 Instantaneous photographs of impinging flame

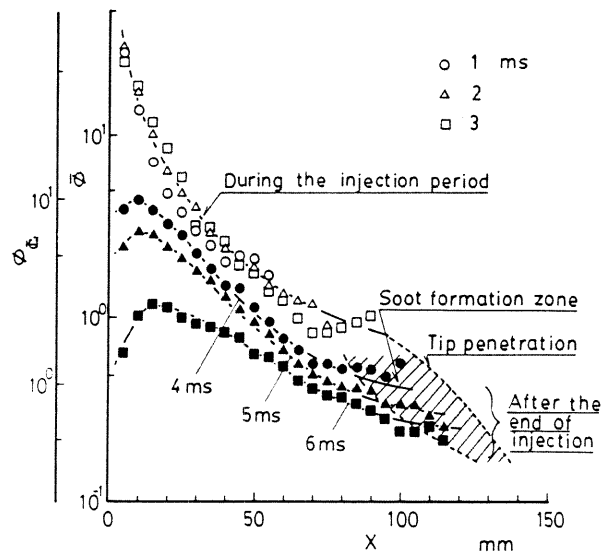


Fig. 13 Equivalence ratio ϕ versus distance X with time as a parameter measured on non-evaporating spray and soot formation zone estimated from instantaneous photographs (ϕ_s : ϕ along spray axis, $\bar{\phi}$: ϕ averaged in cross section perpendicular to spray axis)

Discussions on Mechanism of Formation and Oxidation Processes of Soot Particles in Diesel Flame

It was concluded in the preceding chapter that soot is formed in a zone near the flame tip. There arises a question about the real equivalence ratio in the soot formation zone, because most simulation models assume that soot is formed in a rich mixture zone. A comparison of instantaneous photographs between non-evaporating spray and Flame S in Fig. 7 indicates that difference in tip penetration between them is very small. And our previous paper(13) pointed out that the air entrainment of Flame S is almost the same for non-evaporating spray. These two facts will allow us to assume that non-evaporating spray and flame are very much alike in the distribution of fuel concentration.

Fig. 13 shows the distribution of equivalence ratio along spray axis and its variation with time which were measured by means of an image analysis of high speed Schlieren photographs of a non-evaporating spray(14). $\bar{\phi}$ stands for an equivalence ratio averaged over a cross section perpendicular to the spray axis, while ϕ_s means an equivalence ratio along the spray axis. The hatched zone, which represents the soot formation zone, was drawn from photographs of Flame S in Fig. 7. It is obvious from Fig. 13 that the equivalence ratio in the soot formation zone is almost stoichiometric or less than such. Since PDF of equivalence ratio in the soot formation zone includes a mixture fraction with ϕ larger than 1.5, which is generally known as the sooting limit for hydrocarbon air mixtures, soot in the soot formation zone may be generated in such rich mixtures.

The results and discussions described above can provide a qualitative picture of the formation and oxidation mechanisms of soot particles in the diesel flame. Observing the illustrations shown in Fig. 14, let us summarize and interpret the whole

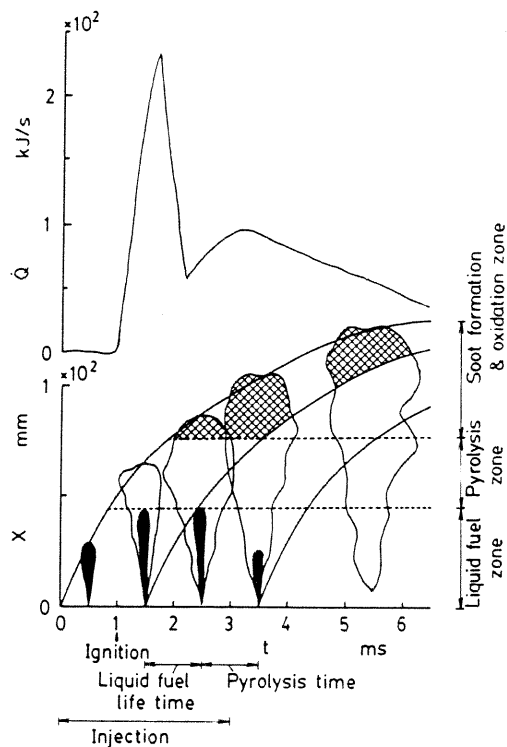


Fig. 14 Illustration of concept of soot formation in diesel flame

sequence of the formation and oxidation of soot particles with the aid of published data available:

1) Ignition delay period - Miwa et al.(15) found in their direct gas sampling study that the fuel which evaporates after traveling a distance of about 40 mm from the nozzle orifice is pyrolyzed rapidly by heat supplied by both hot air and partially reacted combustion products, and that the farther the distance from the orifice is, the higher the concentrations of such pyrolysis products as CH_4 , C_2H_2 , and C_2H_4 become. The authors believe that a similar process is occurring during the ignition delay period of about 1 ms.

2) Initial combustion period - When the cylinder gas pressure starts to increase due to ignition, the spray displays an abrupt increase in volume, except in a region near the orifice, showing rapid heat release throughout the whole spray. The flammable mixture cluster prepared during the ignition delay burns in about 1 ms as a non-luminous flame, and the hot combustion products prevail in the whole flame. Mixture cluster with an equivalence ratio too rich to burn from microscopic view point, undergo pyrolysis since they are exposed to hot combustion products which are dominant in the flame. The rich mixture cluster which has a potential to yield soot particles travel downstream i.e., toward flame tip with the combustion products, as it is being subjected to subsequent pyrolysis, nucleation, and coagulation.

3) Diffusion combustion period - After the lapse of time of around 2 ms from the start of ignition, soot particles appear in a region near the flame tip when the diffusion combustion period is about to start. A gas sampling data taken by Aoyagi et al.(1) with a DI diesel engine shows that soot is detected immediately after the flame tip arrives a sampling position. Our data which exhibit the first appearance of soot particles in a region near flame tip seem to be consistent with their observation.

The fuel injected during this period travels around 40 mm from the nozzle orifice in liquid phase, and is decomposed rapidly by heat as soon as the fuel evaporates. The pyrolysis products penetrate into the burning zone together with the original fuel, as a result, some of them burn and some others are subject to soot formation and of course oxidation processes. When these rich mixture cluster in which soot formation progresses, arrive in a region near the flame tip, soot yield finally amounting to some 20 percent of carbon included in the original fuel. As Fig. 14 shows, if the quasi-steady assumption is made such that each fuel element penetrates in the same manner as the flame tip does, the time required for the fuel vapour to become soot particles will be around 1-2 ms. A gas sampling study with a laminar diffusion flame(16) gives an almost similar picture depicted above.

The dark shadow due to the incident light extinction by soot particles was observed clearly only in a region near the flame tip, while the direct photographs shown in Figures 8 and 10 display that luminous region develops more evenly to a region close to the nozzle orifice although the luminosity is weak as compared to that of the flame tip. This may indicate that soot particles or some soot precursors with low concentration exist in this region. The measurement of flame temperature distribution along flame axis by means of the two-color analysis of color photographs tells a relatively even distribution.

4) Period after the end of injection - The soot

containing region flows downstream after the end of injection, and by 6-7 ms after the start of injection the great majority of soot particles burns out, due to encounter with air through turbulent diffusion.

CONCLUSIONS

The existence of soot particles in a diesel flame in a quiescent atmosphere in a rapid compression machine was visualized by means of the laser schlieren technique. The fuel concentration distribution in a non-evaporating spray which was measured by means of an image analysis technique was used to estimate the equivalence ratio in the soot formation zone of the flame. The temporal and spatial variation of soot concentration in the flame was also correlated with the rate of heat release. A qualitative interpretation of the formation and oxidation processes of soot particles in a diesel flame was deduced from these experiments. The conclusions obtained are:

1) Soot particles in a diesel flame appears first in a region near the flame tip when diffusion combustion period starts, and its concentration is a maximum at about the end of injection, then decreases due to oxidation.

2) The reason for soot being formed in a fuel lean region near the flame tip rather than in a fuel rich region is that the evaporated fuel requires time to be pyrolyzed as it travels through the burning fuel rich zone towards the flame tip.

3) Impingement promotes mixing, and this leads to a higher rate of heat release, a lower soot concentration and a shorter combustion period as compared to a combustion without impingement.

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