KSME International Journal, Vol. 16 No. 6, pp. 851~860, 2002

A Study on the Characteristics of Soot Formation and Oxidation in Free Fuel Droplet Array

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In this study, it was attempted to obtain the fundamental data for the formation and oxidation of soot from a diesel engine. Combustion of spray injected into a cylinder is complex phenomenon having physical and chemical processes, and these processes affect each other. There are many factors in the mechanism of the formation and oxidization of soot and it is necessary to observe spray combustion microscopically. In order to observe with that view, free fuel droplet array was used as an experimental object and the droplet array was injected into an atmospheric combustion chamber with high temperature. Ambient temperature of the combustion chamber, interdroplet spacing, and droplet diameter were selected as parameters, which affect the formation and oxidation of soot. In this study, it was found that the parameters also affect ignition delay of droplet. The ambient temperature especially affected the ignition delay of droplet as well as the flame temperature after self-ignition. As the interdroplet spacing that means the local equivalence ratio in a combustion chamber was narrow, formation of soot was increased. As diameter of droplet was large, surface area of the droplet was also broad, and hence evaporation of the droplet was more active than that of a droplet with relative small diameter.

Key Words : Soot, Free Fuel Droplet Array, Two-Color Method, KL Factor, Ignition Delay

1. Introduction

In recent years, many experimental studies have been attempted to reduce soot from diesel engines. There are two experimental approaches for the reduction of the diesel soot. One is to improve the spray combustion in a combustion chamber and the other is to filter soot particles (Kamimoto *et al*., 1980). Most of the studies, however, are focusing on the combustion improvement to remove the cause of the soot formation. In methods of combustion improvement, the combustion process of the spray in combustion chamber of diesel engine is treated macroscopically and the quantity of soot related to the variation of combustion conditions is measured and analyzed (Lee *et al*., 1997; Lee *et al*., 1996). The influence of the spatial non-uniformity of droplet distribution, the local distribution of temperature, droplet size, and so forth on the soot formation and oxidation are not clarified. Therefore, the mechanism of formation and oxidation of soot are not yet sufficient. The combustion of fuel spray injected into the combustion chamber is a complicated phenomenon in which physical and chemical processes proceed simultaneously with affecting each other

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(Sangiovanni *et al.*, 1984). Therefore, there are many factors in the mechanism of formation and oxidation of soot. In this point of view, it is necessary to provide the fundamental information for the reduction of soot from the diesel engine by observing free fuel droplets and examining the influence of the ambient temperature, droplet size, local section flame temperature, and so forth. In this study, it was attempted to obtain the fundamental data for the formation of soot in free fuel droplet array using two-color method. The flame temperature and KL factors characterizing the qualitative concentration of soot are measured by two-color method with variable conditions of the ambient temperature, droplet diameter, and the spacing between each droplet. The diameter of the free fuel droplets injected into a high temperature gas stream is about 140 μm~160 μm which are obtained by a droplet generator using a piezoelectric transducer.

2. Theory

In the two-color method, the thermal radiations at two different wavelengths are detected so that the flame temperature is then determined from their ratio by eliminating an unknown factor. Intensity of radiation from a black body varies with wavelength and it depends on the temperature of a black body. This is described by Planck's equation

$$E_b(\lambda T) = C_1/\lambda^5(\exp(C_2/\lambda T) - 1)^{-1} \quad (1)$$

The Equation can be described by Wien's equation in visible wavelength.

$$E_{b\lambda}(T) = C_1/\lambda^5(\exp(-C_2/\lambda T)) \quad (2)$$

where $E_{b\lambda}$: monochromatic emissive power of a black body at T, [W/m^3]

λ : wavelength (μm)

T : temperature (K)

C_1 : the 1st Planck's constant $=3.7418\times10^{-16}$ (Wm^2)

C_2 : the 2nd Planck's constant $=1.4388\times10^{-2}$ (mK)

In practice, emissivity; ε_λ has been estimated for soot particles by the widely used empirical correlation of Hottel and Broughton (1932),

$$\varepsilon_\lambda = 1 - \exp(-KL/\lambda^a) \quad (3)$$

where K : an absorption coefficient=proportional to the number density of soot particles

L : the geometric thickness of the flame along the optical axis of the detection system

The value of the parameter depends on the physical and optical properties of the soot in the flame. From Eq. (3),

$$KL = -\lambda^a \ln[1 - \exp(C_2/\lambda(1/T - 1/T_a))] \quad (4)$$

The unknown product KL can be eliminated by rewriting the above equation for two specific wavelengths, λ_1 and λ_2:

$$\begin{aligned} &\lambda_1^{a1}\ln[1-\exp(C_2/\lambda_1(1/T-1/T_{a1}))] \\ &=\lambda_2^{a2}\ln[1-\exp(C_2/\lambda_2(1/T-1/T_{a2}))] \end{aligned} \quad (5)$$

The actual flame temperature T is independent of wavelength, while the apparent temperature T_a and, possibly, the parameter a vary with wavelength. It can be seen that this equation can be solved for the flame temperature T, provided the apparent temperature T_{a1} and T_{a2} for the flame are known at the two wavelength λ_1 and λ_2. These two apparent flame temperatures can be measured at these two wavelengths using a calibrated two-color optical pyrometer system. In fact, this is the purpose of the two-color system: to provide instantaneous measurements of the apparent temperatures T_{a1} and T_{a2} for the flame at two chosen wavelengths λ_1 and λ_2.

3. Experimental Apparatus and Methods

Figure 1 shows the diagram of experimental apparatuses for the measurement of flame temperature and KL factor. The apparatus includes droplet generator system, combustion chamber system, and optical measurement system using two-color method.

The droplet generator used in this study was originally developed by Honoki *et al.* (1985). A cross section of the droplet generator is shown in Fig. 2. The generator includes a piezoelectric

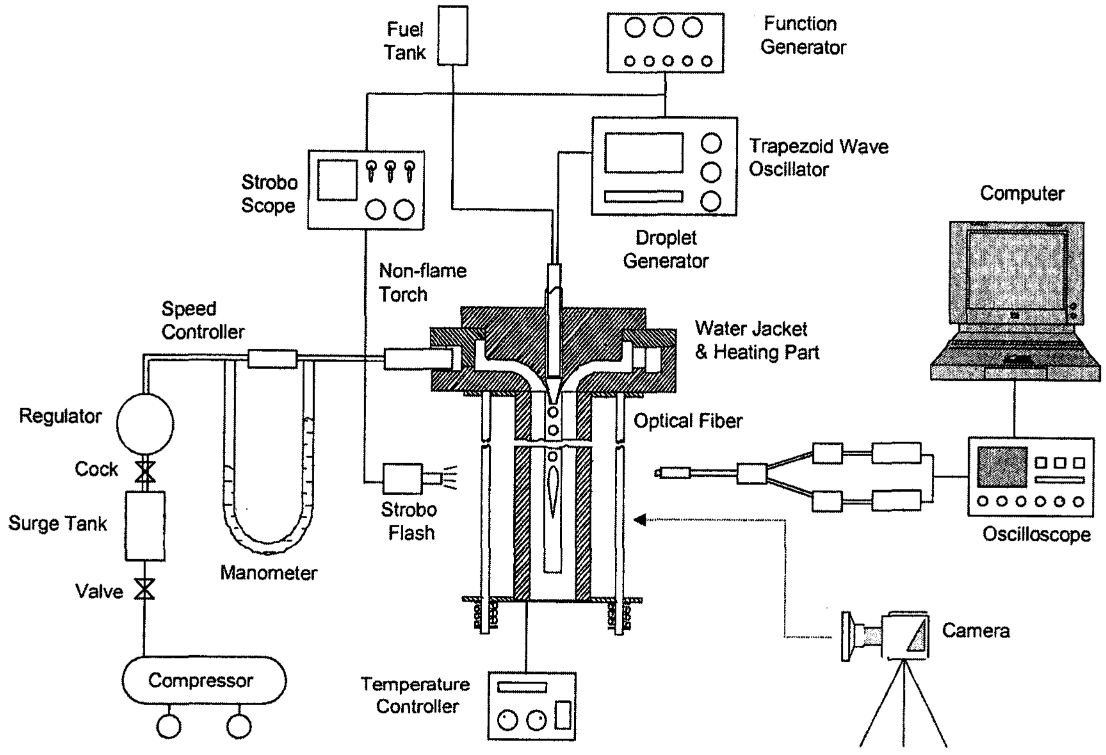

Fig. 1 Schematic diagram of experimental apparatus

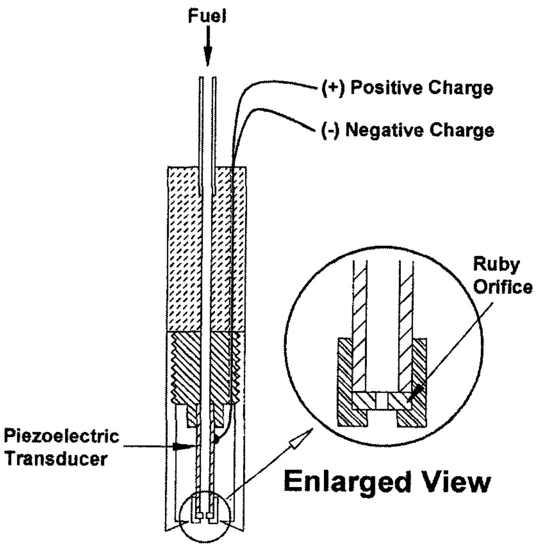

Fig. 2 Diagram of droplet generator

transducer which is made of ceramic of $Pb(Zr, Ti)O_3$ that has the characteristic of contraction when an electric charge is given. A ruby orifice is installed in front of it. It generates a fine droplet with wide ranges of diameter, velocity, and interdroplet spacing. The relative standard deviations of droplet diameter and velocity from their mean values are less than 3%. Test fuel from a fuel tank is transmitted to the transducer through a pipe. An oscillator transforms the rectangular wave into trapezoidal wave and transmits to the transducer. Intermittent signals force the piezoelectric transducer to contract to the radial direction intermittently, and then a generated pressure wave pushes out the liquid through the orifice. (Chung et al., 1990; 1995)

A schematic diagram of combustion chamber used in this study is shown in Fig. 1. The chamber consists of preheating part, heating part, and measurement part. The heated air with temperature of about 773 K is passed to heating part through the non-flame torch of the preheating part. The ambient air from preheating part is further heated here and provided to the measurement part along radial symmetric flow path. The droplet generator is located in the center of heating part. The measurement part is a device to measure the ignition and combustion process of droplet array optically. Quartz windows having good optical transmittance and heat resistance are installed to observe flame of droplet array and to measure soot concentration in the both sides. In the measurement part where droplet array is burned, two-color method is used to obtain the fundamental information about the process of soot formation and oxidation and flame temperature of droplet array using optical fiber. The instruments of two-color method consist of an optical probe with optical fiber, two band-pass filters, photo-diode (Hamamatsu S2281-01), and photo-sensor amplifier (Hamamatsu C2719). Two band-pass filters are used to take chosen wavelengths, 550 nm and 755 nm at center wavelength, in the flame (Lee *et al.*, 1996).

The ambient temperature, droplet diameter, and droplet spacing to be used as the parameters and the corresponding values in this study are shown in Table 1. Because the ambient temperature has influence on the evaporation of fuel in a diesel engine, the characteristics of soot formation is different with the ambient temperature. The temperatures of 973, 1073, and 1173 K are used as the test temperatures to obtain information for the formation and oxidation of soot with the ambient temperature of a combustion chamber.

The number density distribution of fuel droplet is a significant parameter in diesel combustion, because the mixture states of fuel and air have influence on the degree of formation and oxidation of soot. By setting droplet spacing as a parameter in this study, therefore, it is attempted to find influences of the utilization coefficient of air with distance between each droplet on the characteristics of soot.

In an actual diesel engine, it is attempted to increase the surface area of fuel by atomization of fuel as much as possible. After evaporating fuel as fast as possible, subsequently it is attempted to burn fuel completely through accomplishment of homogeneous mixture. Therefore, droplet diameter is to be chosen as a parameter of this study, and it is tried to know how the quantity of fuel-vapor for variation of droplet surface has influence on the characteristics of soot.

Table 1 Experimental parameters and values

Parameter	Values
Ambient temperature, (K)	973, 1073, 1173
Interdroplet spacing, (mm)	5.6, 7, 9.3
Droplet diameter, (μm)	140, 145, 160

When the droplet is injected into the measurement part after passing through the path in the cooling jacket, a relative velocity between the ambient and droplet is produced. The relative velocity is required to be low enough to obtain useful data. The air velocity supplied into the measurement part is fixed at 0.7m/s, and the initial velocity of droplet is adjusted to the air velocity in order to minimize relative velocity between air and droplet as much as possible.

In this study, in order to remove third-party factors that have influence on the formation of soot except for the concerned experimental parameters, the test fuels have the same chemical composition. *n-hexadecane* ($C_{16}H_{34}$) whose *alkane* group has the molecular structure of chain link and *a-methylnaphthalene* ($C_{10}H_7CH_3$) whose aromatic hydrocarbon group has the molecular structure of ring link, having the cetane number 50 mixed with the same volumetric ratio. The former usually has the cetane number 100 and short ignition delay owing to good igniting characteristics. However, because the latter has poor ignition characteristics, the cetane number is zero. The cetane number of test fuels is mole fraction of *n-hexadecane* mixed with *a-methylnaphthalene*.

Two pulses from a pulse generator are transferred to trapezoidal wave oscillator and stroboflash, respectively. The pulses are transformed to trapezoidal waves, which are determined by voltage level of the oscillator and transferred to a piezoelectric transducer. A droplet is generated from liquid fuel in the transducer by repetitive contracting operation through an orifice. The generated diameters are 140, 145, and 160 μm, respectively. The interdroplet spacing is controlled by a pulse generator with 75, 100, and 125 Hz, respectively. They correspond to 9.3, 7, and 5.6 mm, in the distance from the center of a droplet to that of the next droplet.

An optical probe of two-color method is made of optical fiber. The optical probe is installed on a transferring bed to move along flame of droplet array, and adhered closely to the quartz window. The scale is attached to the transferring bed to measure the ignition point of droplet array and flame length in combustion chamber. The light of flame is divided into two parts, through two band-pass filters of 550 nm and 755 nm, each of which is changed into electric signal by a photodiode. The changed signal is saved by a computer after amplified by a photo-sensor amplifier. The droplet array forms thin flame downward from ignition point in the combustion chamber. The optical probe is moved from the ignition point of flame to the end point of combustion by the transferring bed, and the electric signals are measured and saved by an oscilloscope and a PC. A series of these processes are repeated ten times at the same point of flame continuously. The measured voltages are input to a program and calculated into KL factor and flame temperature by a program.

Figure 3 shows the schematic diagram of measurement method. Injected droplet array into the measurement part with high temperature is ignited by itself after some ignition delay. In order to measure KL factors for time history of droplet array flame, with dividing the measurement position of optical probe by 10 mm, they are measured from ignition point to the end of flame. Although two-color method to be used in this study is not able to classify and analyze the process of soot formation and oxidation, it can get qualitative values of variation in soot concentration with time history. Hence, it is necessary to measure the KL factors of flame as above and to divide into the beginning, the middle, and the end period for the flame length, which are compared with factors of each period relatively. Therefore, it comes to obtain the information for formation and oxidation of soot with KL factors in the period.

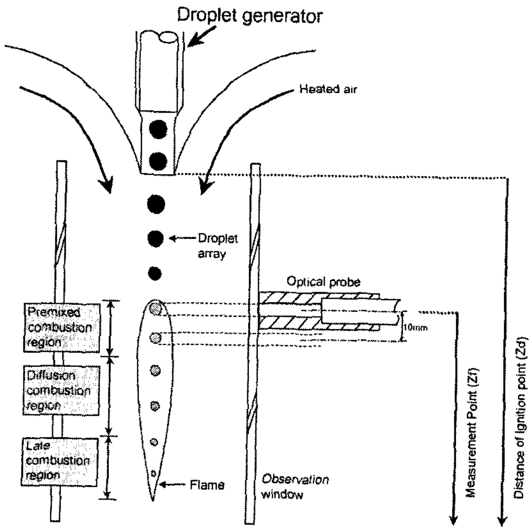

Fig. 3 Diagram of the optical measurement method

4. Results and Discussion

4.1 Ignition delay in each condition

Figure 4 shows the distance between injection entrance and ignition point of droplet array with ambient temperature in interdroplet spacing, 5.6 mm, diameter of droplet, 160 μm. As the ambient temperature increases, the ignition point is advanced to the entrance. The ignition time in the ambient temperature 1173 K is earlier than that in 973 K. Therefore, the higher ambient temperature around droplet is, the more appropriate the condition for self-ignition of droplet is. Figure 5 shows the distance between injection entrance and ignition point of droplet array with interdroplet spacing in ambient temperature, 1173 K, diameter of droplet, 160 μm. As interdroplet spacing increases, the time of ignition is delayed more and more. The narrower space between droplets is, the denser fuel vapor around droplet is, so thet droplet array has proper condition for self-ignition early. Figure 6 shows the distance between injection entrance and ignition point of droplet array with the variation of droplet diameter in ambient temperature, 1173 K, interdroplet spacing, 5.6 mm. As droplet diameter increases, the time of ignition is delayed more and more. The smaller the diameter of droplet is, the denser fuel vapor around droplet is, and hence droplet array has proper condition for early self-ignition.

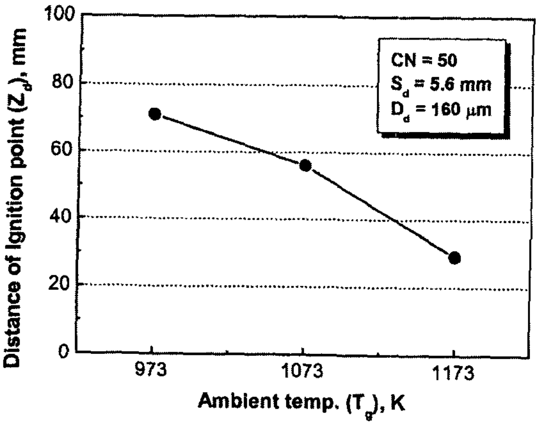

Fig. 4 Distances of ignition point with ambient temperature

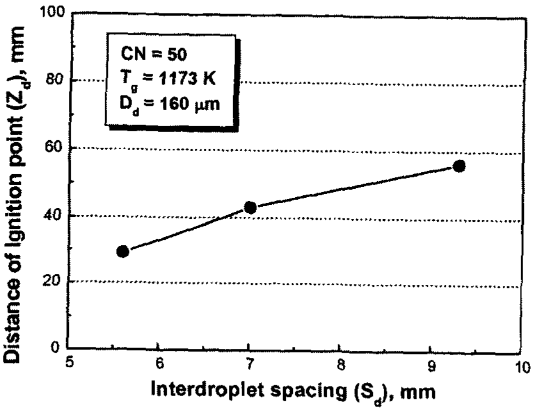

Fig. 5 Distances of ignition point with interdroplet spacing

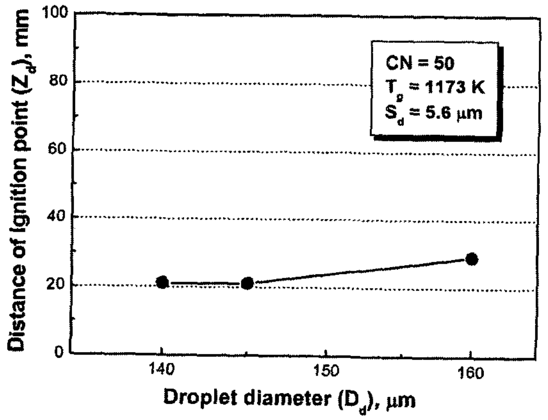

Fig. 6 Distances of ignition point with droplet diameter

4.2 Influence of the ambient temperature on the soot formation

Figure 7 shows flame temperature and *KL* factors from ignition to the end of combustion of droplet array with mean values of experimental results of five times. The experimental conditions are cetane number 50, droplet diameter, 160 μm, interdroplet spacing, 5.6 mm. As ambient temperature increases from 973 K to 1173 K in Fig. 7, flame length of droplet array is about 40, 50, and 60 mm, respectively. As ambient temperature decreases, because the ignition delay increases, the maximum temperature of flame is moved to the ignition point of flame.

Before droplet array is ignited by itself, quantity of fuel vapor depends upon the ignition delay. Hence, flame temperature is high because of the large number of fuel vapor. After ignition, however, because droplet is small relatively to the case of short ignition delay, flame temperature decreases gradually because of a little amount of

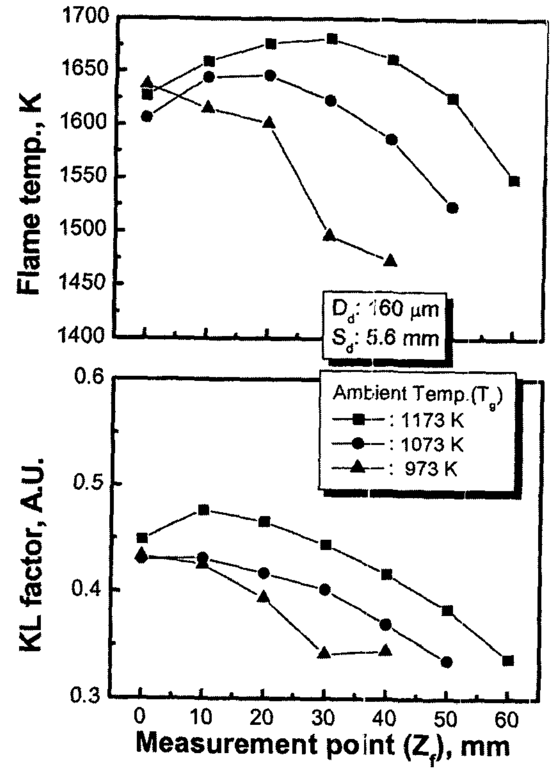

Fig. 7 Flame temperature and KL factors with the ambient temperature

fuel vapor. On the other hand, in the case of short ignition delay, droplet is large on the ignition time relatively to the case of long ignition delay. Because droplet array in the flame has high temperature environment, and the quantity of fuel vapor increases and flame temperature increases gradually with the progression of combustion. After then, since the fuel vapor quantity of droplet is small, it is considered that flame temperature decreases gradually. In all conditions of ambient temperature, *KL* factors are generally high at the early combustion phase and then gradually decrease, *KL* factors have almost similar values with each other at the late combustion phase. In the case of 1173 K, *KL* factors are higher than those of other conditions. *KL* factors are generally proportional to the gradient of flame temperature, and hence ambient temperature has influence on the *KL* factor. Therefore, ambient temperature has influence on the ignition time and evaporation quantity of fuel, accordingly these influences determine premixed quantity and flame temperature of combustion duration. After ignition of droplet, flame temperature has influence on the *KL* factor, and hence it is considered that flame temperature might leads to formation and oxidation of soot.

Figure 8 shows mean value of *KL* factors for the variation of temperature in the case of premixed, diffusion, and late combustion phase. *KL* factors have low values in the case of 973 K and 1073 K, and formation of soot is relatively larger than the condition of 973 K and 1073 K in the case of 1173 K. In the case of 1173K, *KL* factors are higher by about 8% than those of 973K, 1073K in the phase of premixed combustion. As the ambient temperature increases, *KL* factors increase in the phase of diffusion combustion. In the phase of late combustion, *KL* factors are high in the condition of 1173 K. From these results, high ambient temperature results in the increment of soot formation. It is considered that soot formation increases because the amount of diffusion combustion is relatively large due to the shorter ignition delay. Soot is oxidized after ignition, however, quantity of oxidation is the largest in the case of 1173 K. Therefore, in order to reduce soot, it is necessary to increase the quantity of premixed mixture to be burned in the early combustion phase, and to raise the flame temperature in the late combustion phase.

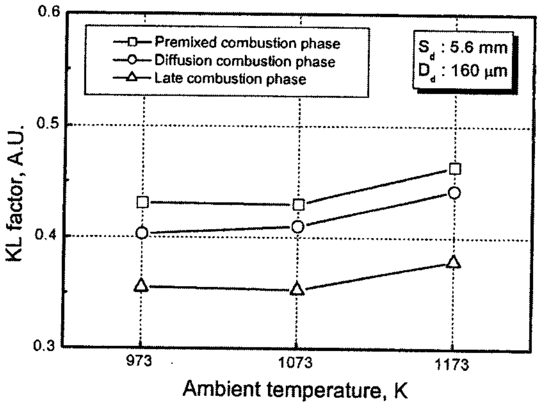

Fig. 8 Relation between KL factors and combustion phase with the ambient temperature

4.3 Influence of the equivalence ratio of local area on the soot formation

In order to investigate the influences of local equivalence ratio on the formation of soot, flame temperature and KL factors were measured from the ignition of droplet to the end of combustion and Fig. 9 shows the results. The experimental conditions were cetane number 50, droplet diameter, 160 μm, and ambient temperature, 1173 K. Then the interdroplet spacing was varied with 5. 6 mm, 7 mm, and 9.3 mm. The figure shows mean values of experimental results of five times.

As interdroplet spacing is reduced from 9.3 mm to 5.6 mm, the initial flame temperature is reduced from 1661 K to 1627 K. It is shown that the occurrence of maximum temperature of flame moves from the initial part to center part of flame. In the results of characteristics of ignition delay considered in the former section, as interdroplet spacing increases, it is ascertained that the ignition time is delayed. According to these tendencies, fuel vapor from droplet is accumulated gradually during the ignition delay, so that fuel vapor has influence on each other. Hence, because quantity of fuel vapor increases as interdroplet spacing decreases, rich equivalence ratio region is formed locally. Although the ignitable mixture is

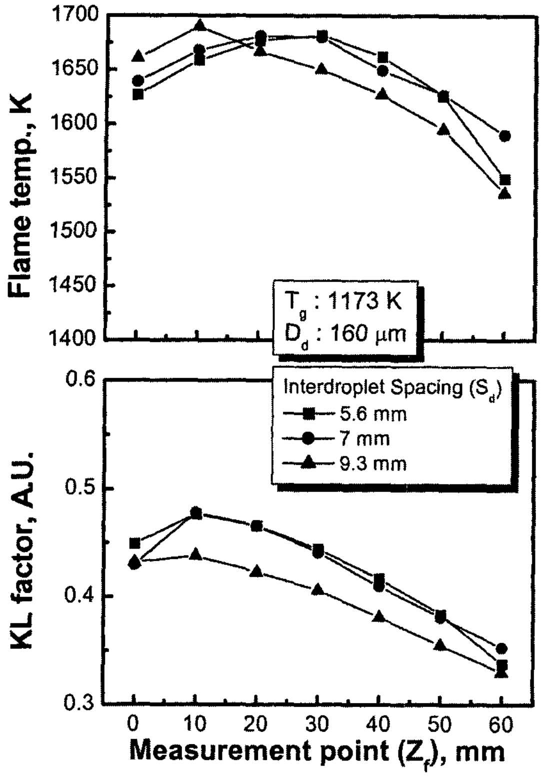

Fig. 9 Flame temperature and KL factors with the interdroplet spacing

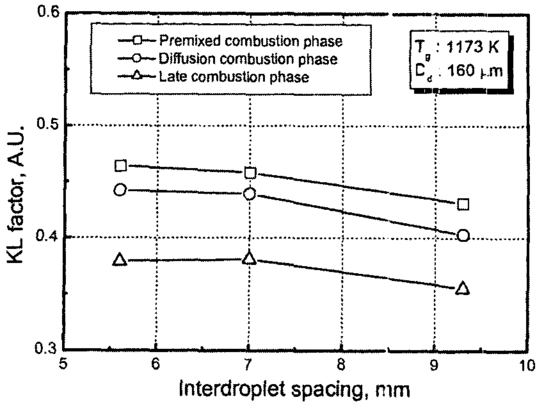

Fig. 10 Relation between KL factors and combustion phase with the interdroplet spacing

relatively formed early so that the ignition time is advanced, flame temperature is low because contact degree with ambient air is relatively narrow. As combustion progresses, because droplet diameter decreases, fuel vapor gets out of the influence of mutual overlap as detailed above, maximum temperature of flame moves to central part of flame gradually. *KL* factors in initial phase of flame show high values and get lower gradually. Especially, regardless of flame temperature, *KL* factors are low in case of 9.3 mm. On the other hand, *KL* factors and flame temperature are similar in the case of 5.6 mm and 7 mm. From the above results, in narrow interdroplet spacing, that is, when local equivalence ratio is rich, high flame temperature has influence on the soot formation. However, the utilization coefficient of air is more dominant than flame temperature in the case of 9.3 mm. Therefore, soot is formed largely in locally rich equivalence ratio region and high flame temperature. As interdroplet spacing is wide, the utilization coefficient of air is thought to be more dominant than flame temperature, and hence soot is oxidized largely.

Figure 10 shows mean value of *KL* factors with variation of interdroplet spacing in case of premixed, diffusion and late combustion phase. As interdroplet spacing increases, formation of soot decreases and final quantity of soot is also low. As interdroplet spacing gets narrow, utilization coefficient of air is low and flame temperature is high because the equivalence ratio is rich. Then, formation of soot increases through thermal pyrolysis of fuel. On the other hand, formation of soot is relatively low and final quantity of soot is low in case of 9.3 mm.

4.4 Influence of droplet size on the soot formation

Figure 11 shows flame temperature and KL factors for variation of droplet diameter in ambient temperature, 1173 K, interdroplet spacing, 5.6 mm, and cetane number, 50. As droplet diameter increases from 145 μm to 160 μm, flame length grows longer from 6 mm to 15 mm and flame temperature is also high. The slope of flame temperature is similar, and maximum flame temperature occurs at middle point.

As droplet diameter is small, *KL* factors are high in the initial part of flame. Subsequently, it means that soot is oxidized rapidly. Hence, oxidation rate of soot is inversely proportional to the droplet diameter. Evaporation and fuel vapor

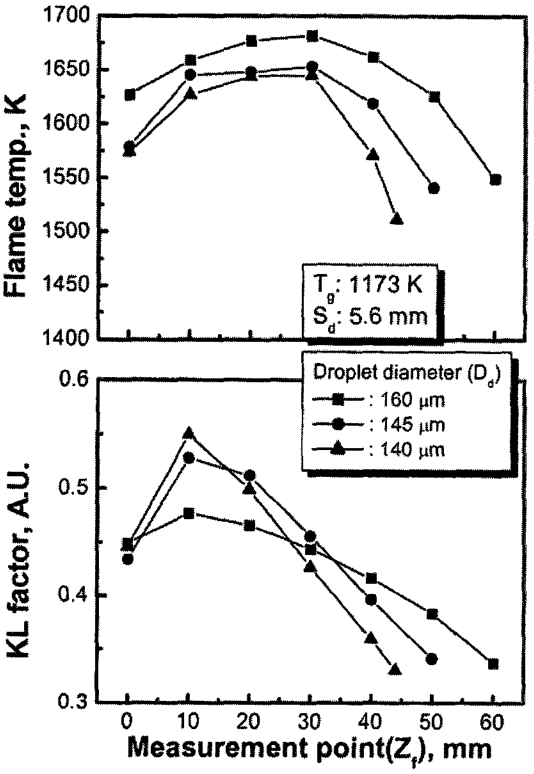

Fig. 11 Flame temperature and KL factors with the droplet diameter

around droplet array increase because surface area of large droplet is wider than that of small droplet. Therefore, flame temperature is high in comparison with small droplet when droplet is ignited. *KL* factors are low because oxidation of soot is promoted. On the other hand, in case of small droplet diameter, formation of soot increases in the initial stage, and decreases rapidly after then. It is considered that high *KL* factors are shown because flame temperature is low, so that quantity of diffusion combustion is larger than that of premixed combustion. After middle point of flame, flame temperature is high in each condition and *KL* factors have similar values. However, in the end of flame, *KL* factors are proportional to flame temperature and diameter. As droplet diameter increases, the quantity of soot formation is larger than that of soot oxidation.

Figure 12 shows mean value of *KL* factors for variation of droplet diameter in the case of premixed, diffusion and late combustion phase.

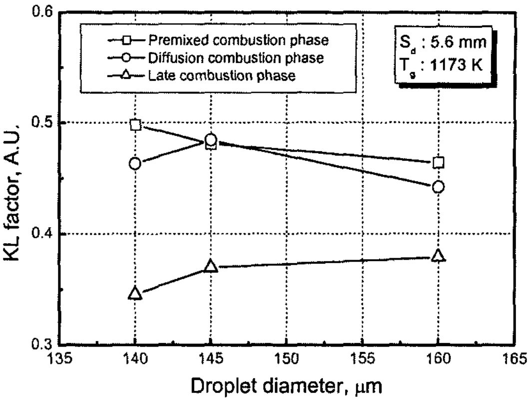

Fig. 12 Relation between KL factors and combustion phase with the droplet diameter

As diameter of droplet increases, *KL* factors decrease in premixed and diffusion combustion phase. However, in late combustion phase, the condition of relatively small diameter results in low *KL* factors. Although initial *KL* factors have higher values than other conditions in the case of small droplet, the *KL* factors show the highest oxidation rate because of low final *KL* factors.

Although the difference of ignition delay is about 2% in three conditions, that of surface area is about 31%. Therefore, it is considered that differences of fuel evaporation and heat transfer through surface area dominate flame temperature and quantity of diffusion combustion. Therefore, increment of surface area of fuel promotes the evaporation and heat transfer of fuel in diffusion combustion and increment of the utilization coefficient of air have influence on the reduction of soot.

5. Conclusions

(1) As the ambient temperature increases, quantity of soot formation is to be large in the combustion of droplet array. The ambient temperature has influence on the quantity of premixed mixture in the early combustion period, which has influence on the flame temperature in diffusion combustion period. Therefore, in order to reduce soot in the diffusion combustion of the droplet array, the early premixed mixture quantity and flame temperature in the late combustion

period have to be increased.

(2) As interdroplet spacing is narrower, a large number of soot is formed relatively in high flame temperature region because of the more active pyrolysis. On the other hand, less soot is formed relatively in the case of wider interdroplet spacing, because the soot formation is dependent upon the coefficient of air utilization. Therefore, in order to reduce soot in the diffusion combustion of the droplet array, it is desirable to increase the coefficient of air utilization of fuel droplet array with introducing the oxidizer into the flame.

(3) As surface area of a large droplet is broader than that of small droplet, the flame temperature is high because of relatively plenty quantity of fuel vapor through the surface area, so that fuel quantity of diffusion combustion period decreases relatively. Hence, as the diameter of droplet is large, KL factors are low in the premixed and the diffusion combustion phase. Therefore, the increase of surface area of fuel causes the increase of heat transfer and fuel evaporation, and hence helps the reduction of soot in the diffusion combustion.

Acknowledgment

This paper was supported by the Research Fund of Dong-A University in 2000. The authors would like to thank their support during this study.

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