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Spontaneous Ignition of Fuel Droplets in Lean Fuel–Air Mixtures

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Experiments have been carried out on the spontaneous ignition of single fuel droplets in lean fuel–air mixtures. The residence time, which is defined as the elapsed time from the introduction of the mixture into the furnace to the insertion of the droplet, is 15 s. The change in the chemical species concentration in fuel–air mixtures before the insertion of the droplet is predicted by the calculation of chemical reaction. In the case of lower ambient temperatures, the methane–air mixture does not react so much and most of methane and oxygen in the ambience survives for the residence time of 15 sec. In this case, the ignition delay time in mixtures is almost the same as that in air, which indicates that the existence of fuel in the ambience has little effect on the spontaneous ignition behavior. On the other hand, the ignition delay time in mixtures becomes larger than that in air for higher ambient temperature. This is mainly due to the decrease in the oxygen concentration in the ambience caused by chemical reaction of fuel–air mixtures during the residence time. However, the quantitative comparison suggests that the increase in the ignition delay time in fuel–air mixtures is not explained by only reduced oxygen concentration.

Introduction

SOME attempts at ignition control of fuel sprays in internal combustion engines such as diesel engines have been made to improve thermal efficiency and reduce pollutant emissions. The prevention of backfire in the prevaporized tube is one of the most important technical subjects for the development of the lean premixed-prevaporized (LPP) combustor for gas turbines. It is well known that understanding of the detailed process of spontaneous ignition of fuel sprays is required to solve problems associated with these subjects.

Because fundamental information of spontaneous ignition of a liquid fuel droplet is indispensable for better understanding the spray ignition mechanism, many studies have been performed on the spontaneous ignition of a fuel droplet suspended at the tip of a fine fiber or a freely falling droplet in high-temperature environments. Pioneering works on droplet ignition[1–4] have been reported from 1960s. The effects of liquid fuel properties,[5,6] ambient pressure and temperature,[7–10] initial droplet diameter,[11] and ambient flow velocity[12] on the ignition process have been explored by analytical and experimental research of fuel droplets. Attention has been also paid to the effect of droplet interaction in dispersed fuels on the ignition process, and experiments on ignition of droplet arrays[13,14] and droplet matrices[15] have been conducted. Experimental results have shown that the ignition delay time has a minimum at a certain spacing of the droplet array, which is consistent with the predicted result. The occurrence of two-stage ignition behavior, which is defined as the appearance of hot flame that follows the cool flame caused by the low-temperature mechanism, has been proved by temperature measurements by thermocouple and interferometer.[16,17] The detailed mechanism of two-stage ignition has been studied experimentally and analytically.[4,6,18,19] It was found that the type of ignition behavior is strongly dependent on the ambient pressure and temperature, and a map of the ignition region on a pressure and temperature plane has been obtained for various n-alkane droplets. A numerical simulation of droplet ignition, including liquid-phase heating,[20] and an unique experiment of droplet ignition by the laminar flame propagating homogeneous fuel–air mixtures[21] have been recently conducted.

From the viewpoint of the spray ignition process in actual combustors, it is considered that droplets in sprays ignite spontaneously in the environment in which there exists prevaporized fuel. It is important to explore the ignition process in such an environment, especially for the understanding of ignition control by pilot injection in diesel engines and backfire in LPP combustors. There have been few studies on the effect of prevaporized fuel in ambience on droplet ignition, except for the analytical study by Niioka et al.[22] They calculated the ignition delay time by using the one-dimensional numerical simulation with an irreversible overall one-step reaction. The calculated result showed that a small amount of fuel vapor, less than 0.5%, has little effect on the ignition delay time for n–heptane droplets. Experimental results are needed to confirm the validity of the prediction in the previous study.

The primary objective of the present work is to obtain experimentally the basic information needed for the understanding of the effect of prevaporized fuel that exists in the ambience on the spontaneous ignition process of a fuel droplet. A suspended fuel droplet is subjected in lean fuel–air mixtures under high-temperature environments, and the ignition delay time is measured. In the experiments, a lean mixture prepared in the mixing tank is introduced into the high-temperature ambience in the electric furnace. The mixture is expected to react before the insertion of the droplet into the furnace and the chemical composition of the ambience to change with time. The zero-dimensional calculation on the chemical reaction of fuel–air mixtures is carried out, and the time history of the chemical composition in the ambience is predicted. Based on the predicted results, effects of the change in the chemical composition in the ambience on the droplet ignition process are discussed.

Experimental Apparatus and Procedure

Figure 1 shows the apparatus for the experiments of spontaneous ignition of fuel droplets, which is almost the same as that employed in previous studies.[5,6] The droplet generating system, the droplet translation system, and the electric furnace are located in the combustion chamber, which has an inner space of 100 mm in diameter and 320 mm in height.

Figure 2 shows the details of droplet generating system and droplet translation system. A suspended droplet is formed at the

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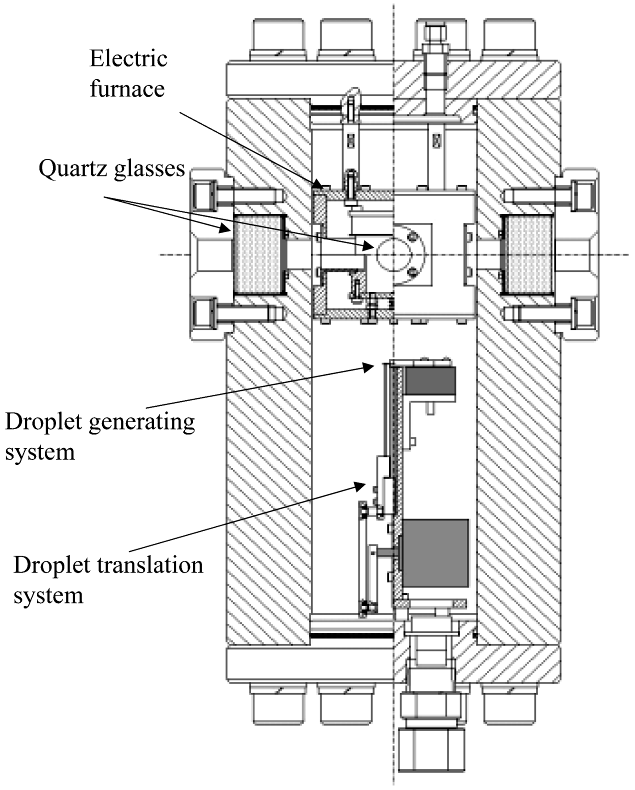

Fig. 1 Schematic of experimental apparatus.

lower section of the combustion chamber. The fuel is fed on the tip of the suspender through the nozzle, the other end of which leads to the fuel tank located outside the chamber. The nozzle is fixed on the arm, which is movable with a rotary solenoid, and it is retracted after the droplet generation to not obstruct the insertion of the droplet into the furnace. The suspender is made of quartz glass fiber with a diameter of 0.15 mm. The suspender is placed horizontally so that it may have a minimum cooling effect on the gas in front of the droplet at its insertion. The formed droplet is gradually heated before the insertion into the furnace because of the existence of the hot electric furnace at the upper section. The droplet temperature is monitored during the preparation of the experiments with a K-type thermocouple with a diameter of 0.05 mm. The hot junction of the thermocouple is fixed on the tip of the suspender, and the temperature is measured in the vicinity of the center of the droplet. The time constant of this thermocouple is estimated to be about 10 ms. In the experiments, the droplet temperature just before insertion into the electric furnace, which is called the initial droplet temperature, is kept constant (412 K ± 5 K). The initial droplet diameter is varied from 0.6 to 1.2 mm. The employed liquid fuel is n–dodecane ($C_{12}H_{26}$), whose boiling temperature is 489.5 K.

The suspended droplet can be quickly inserted into the electric furnace with the droplet translation system controlled by the computer. The droplet suspender is mounted on the sliding rack, which fits with the linear guide. The sliding rack is connected to the crank, which is rotated by the stepping motor. Thus, the rotation of the stepping motor brings the suspended droplet upward into the furnace and again downward. A constant acceleration and deceleration is applied to minimize the required time for the insertion, which achieves the insertion time within 100 ms to travel the distance of about 60 mm.

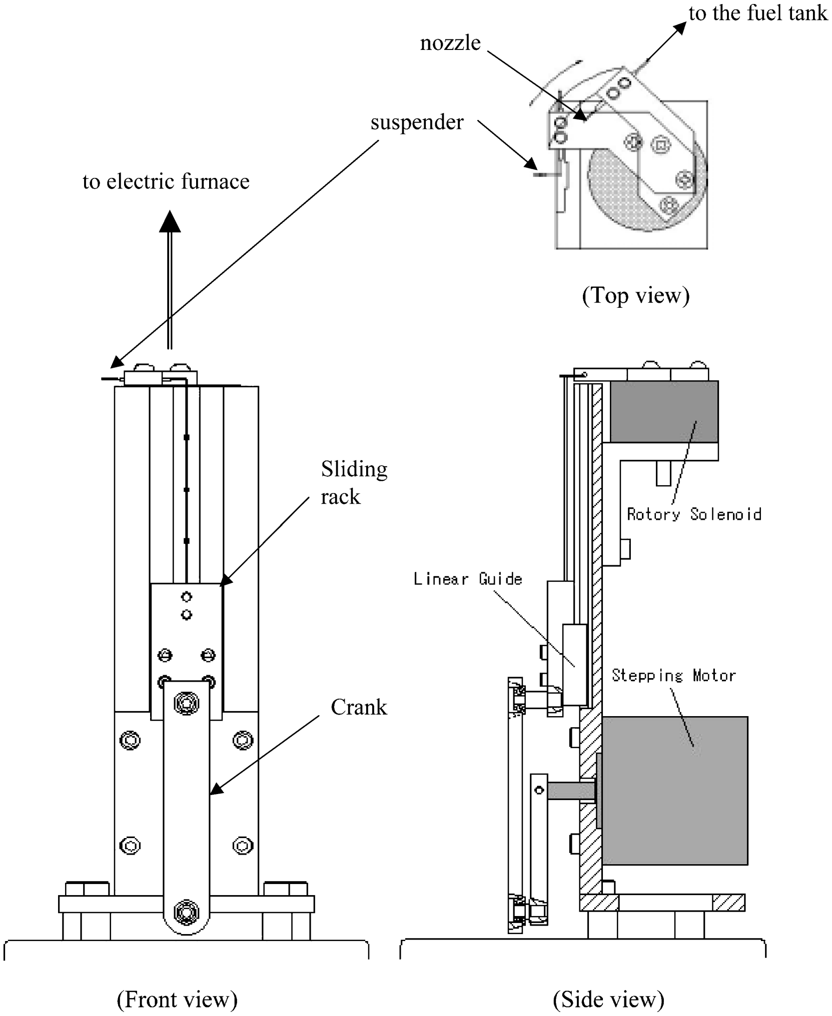

Fig. 2 Details of droplet generating system and droplet translation system.

As shown in Fig. 1, the electric furnace is located at the upper section of the chamber, which is a stainless-steel cylinder with an inner diameter of 40 mm and a height of 50 mm. Four quartz windows are installed on the side wall of the furnace for observation of the ignition behavior. The ambient temperature in the furnace is measured by a K-type thermocouple, with a diameter of 0.1 mm. The electric output from the thermocouple is given as a feedback to the controller, which can automatically control the electric current to keep the ambient temperature as a predetermined value. The thermocouple is located at the same level as the position where the droplet stops after its insertion to reduce the error in measuring the ambient temperature due to the temperature gradient in the vertical direction in the furnace. The time constant of the thermocouple is about 0.2 s, and it is enough for monitoring of the ambient temperature. The influence of the radiative heat loss on the thermocouple measurements is estimated from the correction method proposed by Kaskan.[23] The ambient temperature is determined by calculating the radiation correction. The measured thermocouple temperature is 836 and 918 K, respectively, when the corrected temperature is 873 and 973 K.

The optical system is shown in Fig. 3. The charge-coupled device (CCD) camera with the frame speed of 30 frames/s with a backlight is used to obtain the droplet image, and the initial droplet diameter was estimated from the image. The color high-speed video camera (NAC MEMRECAM ci-Expo) with the frame speed of 500 frames/s and a lens with the focal length of 15 mm is employed for the observation of the luminous flame formed around the droplet at the spontaneous ignition. Another high-speed camera (Photron FASTCAM-ultima SE) with the frame speed of 2000 frames/s is also employed with the installation of the image intensifier for the detailed observation of the OH emission distribution at the droplet ignition. The frame speed for the OH emission observation is set to be 4500 frames/s. The ignition delay time is measured from the measurement of the OH emission by using the optical interference filter with the maximum transmission at 306.4 nm and photomultiplier (Hamamatsu R7449), which has sensitivity to the light whose wavelength ranged from 185 to 650 nm. The analog signal from the photomultiplier is stored in a personal computer with the sampling interval of 1 ms. The ignition delay time is defined as the time period from the time when the droplet becomes stationary in the furnace after its insertion to the time when the OH emission is detected. The uncertainty in measuring the ignition delay time is less than several milliseconds. Observation by the high-speed video camera shows that the time when the luminous flame appears around the droplet is almost consistent with the time when the OH emission is detected.

The fuel–air mixture for the ambient gas is prepared manometrically in a stainless-steel tank, whose capacity is 60 liters, and is mixed by a stirrer before use. To investigate the effect of prevaporized fuel in the ambience on the ignition process of the droplet, n–dodecane, which is the same fuel as the droplet, may be desirable as the fuel for the ambient mixture. For simplicity of the experimental procedure, methane, propane, and n–heptane are employed as fuel for mixtures. Although the use of different fuels for ambient mixture and liquid droplet seems to complicate the interpretation of experimental results, it could be possible to explore qualitatively the effect on the droplet ignition of prevaporized fuel in the ambience from the viewpoint of hierarchy in the reaction mechanism of alkane hydrocarbon fuels.[24,25] The mixtures used in experiments are very lean, and the fuel concentration is out of the lower flammable limit to prevent spontaneous ignition of mixture itself in high-temperature environments. The relationship between the lower flammable limit and the ambient temperature is indicated by the following equation suggested by Zabetakis et al.,[26] which is based on Burgess–Wheeler's law:

$$L_t = L_{298} - (0.75/\Delta H_c) \times (t - 298) \qquad (1)$$

Here, ΔH_c is heat of combustion (kilocalories per mole) and L_{298} and L_t are lower flammable limit (volume percent) at ambient temperatures of 298 K and t (Kelvin), respectively. The lower flammable limit at the predetermined ambient temperature can be estimated from Eq. (1). The fuel concentration is selected to be 0.44% (by volume) and 0.89% for the methane–air mixture, which corresponds

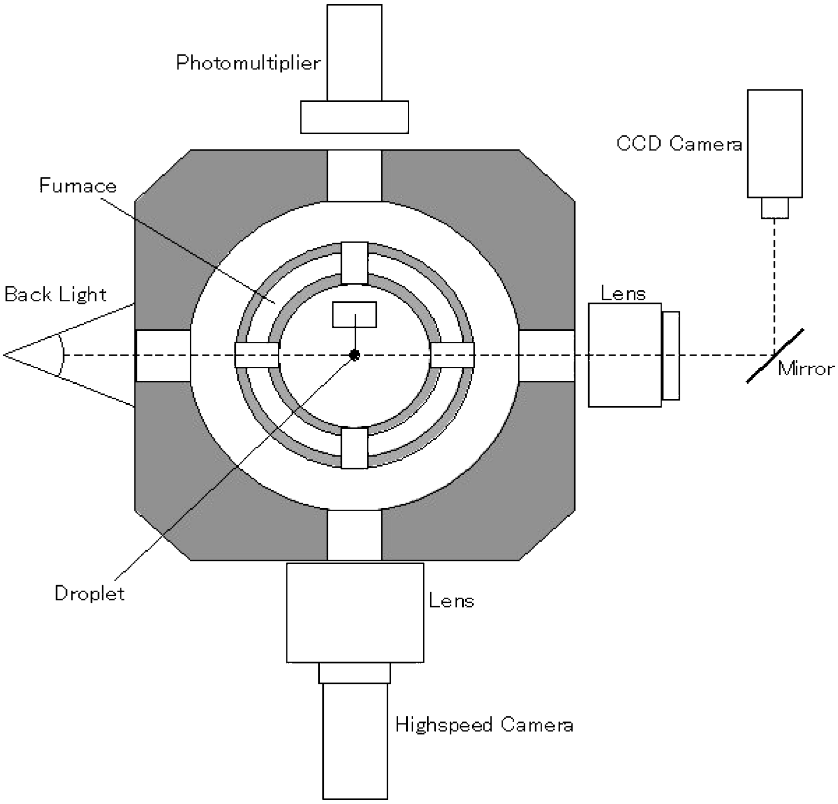

Fig. 3 Schematic of the optical system (top view).

to the equivalence ratio of 0.046 and 0.094, respectively. It is also selected to be 0.75% (equivalence ratio of 0.168) and 0.4% (equivalence ratio of 0.209) for the propane–air mixture and n–heptane–air mixture, respectively. The fuel concentration in the mixtures used in the experiments is confirmed by gas chromatograph measurements. The mixture composition is known to within an error of less than 2%. The oxygen concentration in mixtures is kept to be 21% (by volume) in all cases.

The ambient pressure is 0.1 MPa, and the ambient temperature ranged from 873 to 973 K in the experiments. Because previous studies have reported that the two-stage ignition does not occur in these ambient conditions,[5,17] attention is mainly paid to the appearance of the hot flame, which here is defined as the ignition, caused by the high-temperature reaction mechanism.

Reaction Calculation of Fuel–Air Mixtures

When the fuel–air mixture is introduced into the electric furnace where the temperature is high, the chemical composition is supposed to be changed for the reaction before insertion of the droplet. The change in chemical composition of ambient mixtures in high-temperature environments is predicted by the zero-dimensional calculation of chemical reactions to confirm the ambient gas composition for droplet ignition experiments.

The reaction scheme used in calculations consists of 689 elementary reactions with 155 chemical species, which has been proposed by Koert et al.[27] Calculations are performed using the five-ordered backward differentiation formula (BDF) method (see Ref. 28), which is one of the implicit solution methods.

Figure 4 shows the comparison between experiment and calculation for the time history of the oxygen concentration in 0.6% propane–air mixture for the ambient temperature of 873 K. In the calculations, the temperature of the mixture is assumed to be constant in spite of the heat generation by the chemical reaction. This assumption corresponds to the experimental result that the ambient temperature measured by thermocouple is almost constant after the mixture is introduced into the furnace. The reaction occurs relatively slowly because the mixture is very lean, which may lead to the balance between heat generation by reactions and the heat loss. The oxygen concentration is measured by gas chromatography in experiments. Molecular Sieve 5A is used as the thermal conductivity detector for oxygen. The ambient gas in the furnace is sampled with the sampling probe of inner diameter 2 mm. The scattering of the experimental data is indicated by the vertical line of Fig. 4, which shows that the reproducibility, by repeated sampling on different runs, yielded agreement greater than 90%. The agreement between experiment and calculation is found to be good, which confirms the validity of the prediction of oxygen concentration in the ambient mixture by the reaction calculation.

Figures 5 and 6 show the time histories of methane and oxygen concentrations in 0.89% methane–air mixture for the ambient temperatures of 923 and 973 K, respectively. The results show that methane and oxygen concentrations decrease gradually with time at the initial stage and then they decrease rapidly for both ambient temperatures. Methane is consumed at about 110 s for ambient temperature of 923 K and at about 15 s for ambient temperature of 973 K. The oxygen concentration reduces to about 19.2% after the time when methane is completely consumed for both cases. This residual oxygen concentration is almost the same as that determined from the overall reaction calculation.

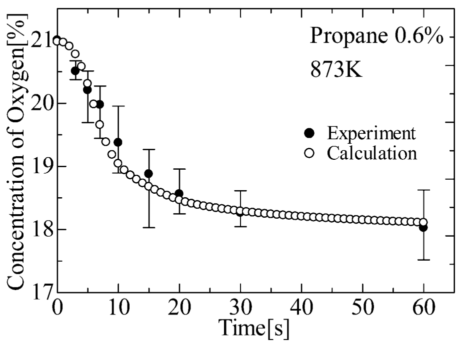

Fig. 4 Comparison between experiment and calculation for time history of oxygen concentration in 0.6% propane–air mixtures for 873-K ambient temperature.

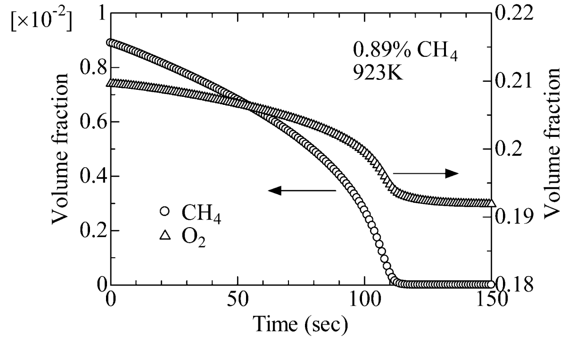

Fig. 5 Calculated time histories of methane and oxygen concentrations in 0.89% methane–air mixtures for 923-K ambient temperature.

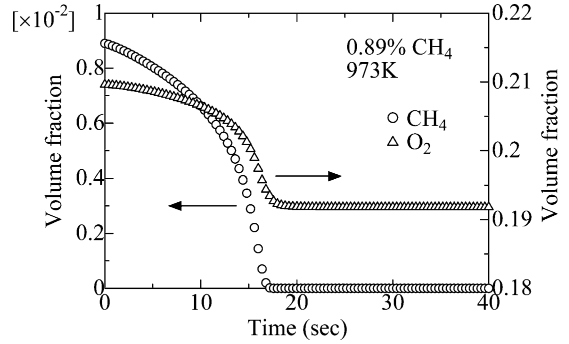

Fig. 6 Calculated time histories of methane and oxygen concentrations in 0.89% methane–air mixtures for 973-K ambient temperature.

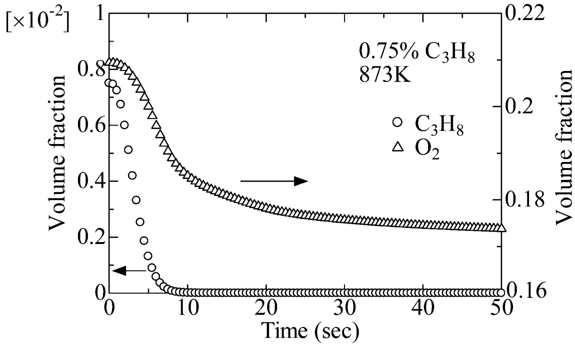

Fig. 7 Calculated time histories of propane and oxygen concentrations in 0.75% propane–air mixtures for 873-K ambient temperature.

Figures 7 and 8 show the results for 0.75% propane–air mixtures at ambient temperatures of 873 and 923 K, respectively. The rapid decrease in propane and oxygen concentrations indicates that the reactivity of propane is much greater than that of methane, which has been well known. Propane is completely consumed at about 10 s for 873 K and at about 5 s for 923 K. The oxygen concentration continues to decrease gradually with time after the consumption of propane, approaching about 17.3%. The decrease in oxygen concentration after the consumption of propane suggests that propane decomposes rapidly to intermediate species, such as small hydrocarbons. Figure 9 shows the concentration profile of representative intermediate hydrocarbons for 0.75% propane–air mixtures for the

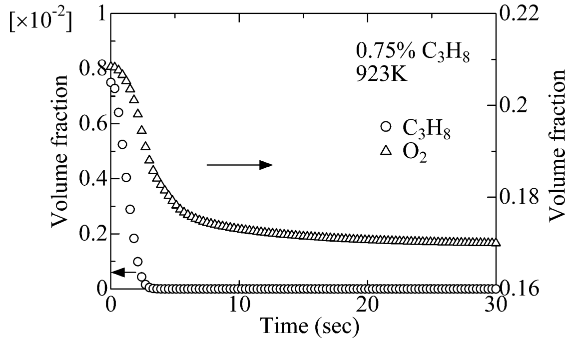

Fig. 8 Calculated time histories of propane and oxygen concentrations in 0.75% propane–air mixtures for 923-K ambient temperature.

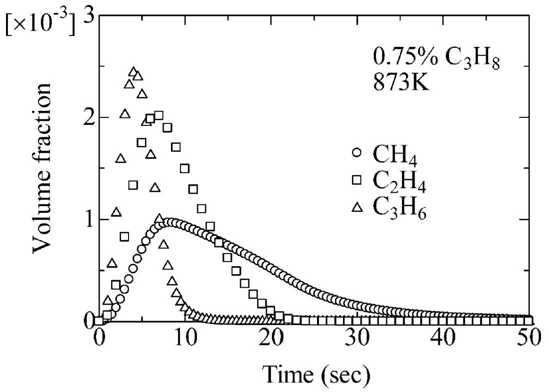

Fig. 9 Calculated time histories of concentration of intermediate small hydrocarbons in 0.75% propane–air mixtures for 873-K ambient temperature.

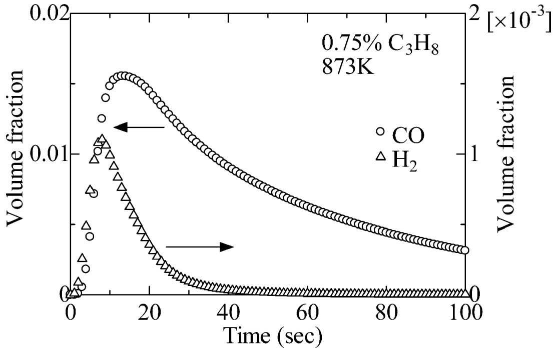

Fig. 10 Calculated time histories of H_2 and CO concentrations in 0.75% propane–air mixtures for 873-K ambient temperature.

ambient temperature of 873 K. The intermediate hydrocarbons are consumed, and H_2 and CO are formed.[29] The H_2–CO oxidation occurs gradually with time, and it continues up to 100 s, as shown in Fig. 10. This propane oxidation behavior is similar to previous results of Biet et al.[30]

Experimental Results and Discussion

The calculated results show that the chemical composition in the ambient gas is changed with time and depends on the residence time of the mixtures in high-temperature environments before the insertion of the droplet. The residence time should be employed as a parameter that indicates the initial chemical composition in ambient gas where a fuel droplet is ignited. It is defined as the elapsed time from the mixture introduction into the combustion chamber to the insertion of a droplet in the furnace, and it is kept to be 15 s for the convenience of the experimental procedure in this work. The procedure for the mixture introduction to the combustion chamber is as follows. First, the combustion chamber is filled with an air and the ambient temperature in the electric furnace is set to a predetermined one. Both the intake and the exhaust valves installed on the combustion chamber are opened and the fuel–air mixture is introduced through the intake valve from the mixture tank to push the residence air through the exhaust valve. The fuel–air mixture continues to flow into the chamber for about 90 s to remove the residence air completely. The introduction of the mixture is finished by closing the intake and exhaust valves. The reduction of the ambient temperature in the furnace occurs during the mixture introduction. Practically, the residence time is determined as the elapsed time from the time when the ambient temperature increases again to the predetermined time of the insertion of a droplet in the furnace. The required time for the recovery of the ambient temperature is several seconds. The oxygen concentration in the fuel–air mixture is measured by a gas chromatograph. The time history of the oxygen concentration in the fuel–air mixture indicates that the fuel–air mixture displaces completely air that was already present in the furnace and that it is unreacted just after the introduction, as shown in Fig. 4. The direct photograph indicates that the droplet flame at ignition becomes almost spherical, as shown in Fig. 11, which suggests that the effect of forced convection caused by the introduction of fuel–air mixture and by the droplet insertion on the droplet ignition process is small.

Figures 12 and 13 show the ignition delay time in methane–air mixtures for ambient temperatures of 923 and 973 K, respectively. These results indicate that the ignition delay time increases as the

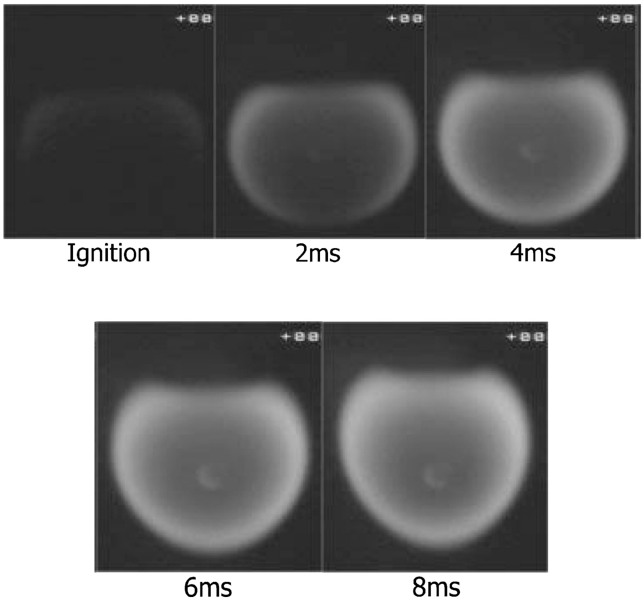

Fig. 11 High-speed camera direct photographs of droplet flame just after spontaneous ignition.

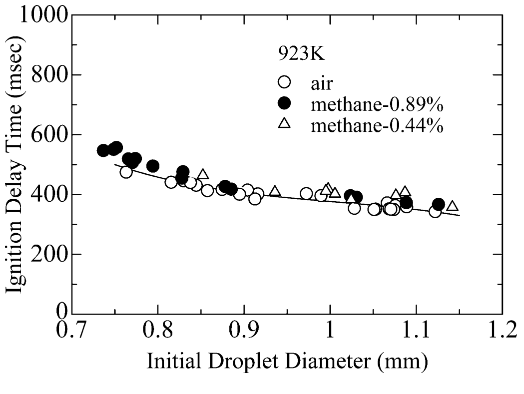

Fig. 12 Ignition delay time in methane–air mixtures for methane concentrations of 0.44 and 0.89%.

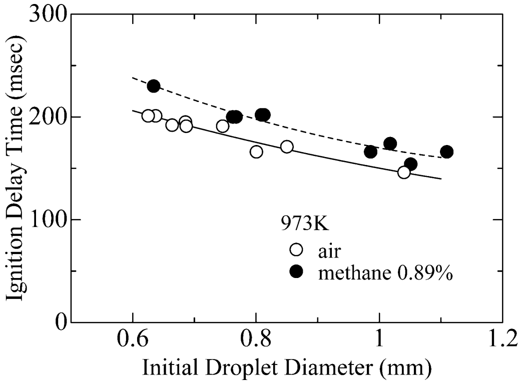

Fig. 13 Ignition delay time in methane–air mixtures for methane concentration of 0.89%.

initial droplet diameter decreases. This tendency is the same as the experimental result[12] for the n–heptane droplet, which has higher volatility than n–dodecane. A high-volatility fuel such as n–heptane produces a large mass flux of the vaporized fuel, and it needs a long reaction time. That is, the vaporized mass flux is inversely proportional to the droplet diameter, and the reaction requires a longer time to consume the large mass flux of the vaporized fuel as the droplet diameter decreases. This results in the increase in the ignition delay time with the decrease in the droplet diameter. That is, the ignition behavior is dominated by the chemical reaction in this case. The previous experimental result[12] shows the opposite tendency for n–decane, which has similar volatility to n–dodecane. This discrepancy is due to the high initial temperature of the droplet in this experiment. Because the initial droplet temperature is relatively high as already mentioned, the n–dodecane droplet easily vaporizes and it produces the large mass flux of the vaporized fuel, although it has lower volatility.

Because little change in methane and oxygen concentrations has occurred at the elapsed time of 15 s for the ambient temperature of 923 K, as shown in Fig. 5, the measured result in Fig. 12 shows that the addition of fuel in the ambient air has little effect on the ignition delay time. This tendency agrees with the analytical result performed by Niioka et al.[22] Although little difference between ignition delay times for air and methane–air mixtures in the smaller initial droplet diameter region is observed, the relative difference to the base ignition delay time for air is only 8% at the maximum. The difference is found to be within 5% in the larger initial droplet diameter region. It is found that the thermal properties of the 0.44 and 0.89% methane–air mixtures, such as specific heat and diffusion coefficient, are almost the same as those of air. This indicates that the addition of a small amount of fuel has little effect on the physical process that is related closely to the ignition process. In addition, the ignition behavior in this experimental condition is dominated by the chemical reaction, mentioned earlier. The reaction related to the ignition process occurs more rapidly than the reaction in the ambient lean mixtures because it is considered that the ignition occurs at the location where the mixture of fuel vapor and air is stoichiometric. Consequently, the additional fuel in the ambience itself plays an unimportant role in chemical process for the droplet ignition, and it behaves as an inert gas.

As shown in Fig. 13, the ignition delay time in 0.89% methane is larger than that in air at the ambient temperature of 973 K. The increase in the ignition delay time is about 20% at the initial droplet diameter of 0.62 mm and is about 15% at the initial droplet diameter of 1.0 mm. In this case, the methane is completely consumed, and the oxygen concentration decreases at the elapsed time of 15 s, as shown in Fig. 4. This indicates that the increase in the ignition delay time may be mainly attributable to the decrease in the oxygen concentration in the ambience by the chemical reaction. It has been reported in previous study[10] that the ignition delay increases with the decrease in the oxygen concentration.

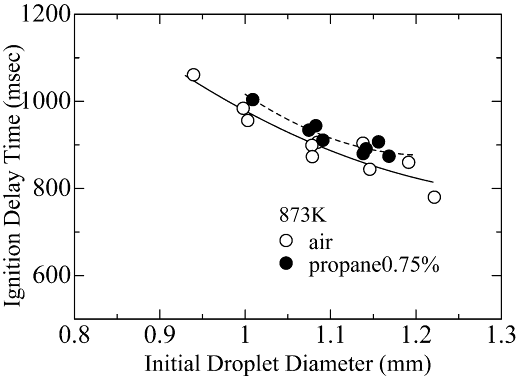

Fig. 14 Ignition delay time in propane–air mixtures for propane concentration of 0.75%.

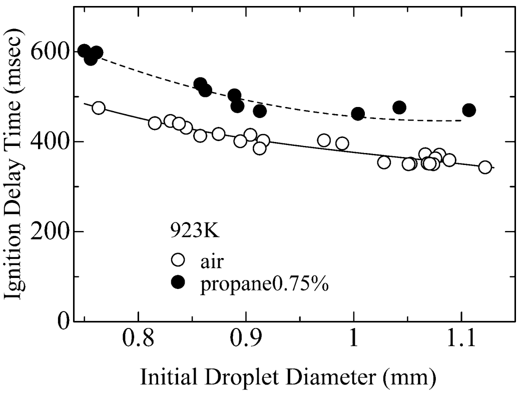

Fig. 15 Ignition delay time in propane-air mixtures for the propane concentration of 0.75%.

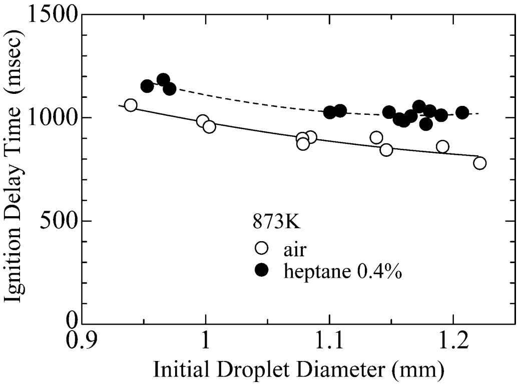

Fig. 16 Ignition delay time in n–heptane–air mixtures for n–heptane concentration of 0.4%.

Figures 14 and 15 show the ignition delay time in 0.75% propane–air mixtures for ambient temperatures of 873 and 923 K, respectively. The ignition delay time in the propane–air mixture is larger that that in air for both cases, which may be due to the decrease in the oxygen concentration in the ambience, as shown in Figs. 7 and 8. It is also found that the difference between ignition delay times in air and mixture for 873 K is not as significant, compared with the result for 923 K. That is, about 25% decrease in ignition delay time for 923 K is observed, whereas only about 5% decrease for 873 K is observed. Figures 7 and 8 show that the residual oxygen concentration in the ambience at the residence time of 15 s is a little larger for 873 than 923 K, which may be one of the reasons for the small increase in the ignition delay time at 873 K.

Figure 16 shows the ignition delay time in n–heptane–air mixtures for the ambient temperature of 873 K. The ignition delay time in the mixture is larger that that in air, which is similar to the results for propane–air mixtures. The time histories of the oxygen concentration in n–heptane–air mixtures are not predicted because the reaction scheme employed in this work is not available for the n–heptane reaction system. It is well known that the reactivity of n–heptane is similar to that of propane, and the assumption that the reduction of oxygen concentration in the ambience occurs at the residence time of 15 s is reasonable. Thus, it is considered that the increase in the ignition delay time in the mixture is due to the decrease in the oxygen concentration.

The effect of the existence of the fuel in the ambience on the droplet ignition can be revealed only in the methane–air mixture in this experiment. The small amount of methane has little effect on the ignition delay time of the droplet, as shown in Fig. 12. It is supposed that this result is due to the low reactivity of methane. For propane, which has relatively high reactivity, the predictions in Figs. 7 and 8 show that the required time for the decomposition in the lean mixture is several seconds, which is much longer than the ignition delay time of a droplet. It is reasonable that the small amount of propane has little effect on the ignition delay time of the droplet, similar to methane. A proper experimental study will be needed to verify this explanation. Also note that the ignition behavior is controlled by the reaction in this experiment, which is revealed by the dependence of the ignition delay time on the initial droplet diameter. The physical process, such as the vaporization, is not considered to affect the ignition behavior in this reaction-controlled region. The ignition behavior is controlled by the vaporization, which has been defined as the vaporization controlled[7,13] for droplets with lower volatility or larger diameter. In this vaporization-controlled region, it is probable that the ignition behavior is affected by the existence of the fuel. Further studies also will be needed in the vaporization-controlled region.

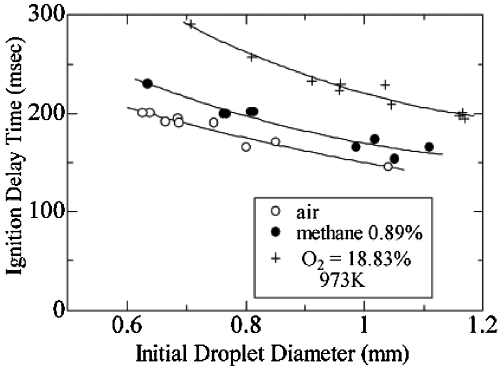

Fig. 17 Ignition delay time for air, 0.89% methane–air mixture and reduced oxygen environment for 973-K ambient temperature.

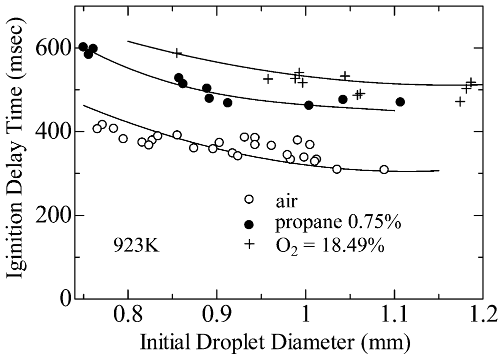

Fig. 18 Ignition delay time for air, 0.75% propane–air mixture and reduced oxygen environment for 923-K ambient temperature.

Figures 13–16 indicate the ignition delay times in the reacted fuel–air mixtures, which are larger than those in air. The increase in the ignition delay time can be explained by the reduction of the oxygen concentration in the ambience, but the explanation is highly qualitative. The quantitative discussion on the effect of oxygen concentration on the ignition delay time is conducted subsequently.

Figure 17 shows the ignition delay time for the ambient oxygen concentration of 18.83% in the nitrogen–oxygen mixture, which is almost the same as the residual oxygen concentration in 0.89% methane–air mixture at the elapsed time of 15 s. Although the predicted result shows that the residual oxygen concentration is about 19.2%, the oxygen concentration in the prepared nitrogen–oxygen mixture is found to be slightly lower by gas chromatography measurements. The ignition delay time in air (21% oxygen concentration) is also indicated in Fig. 17. The ignition delay time in the ambient oxygen concentration of 18.83% is about 1.5 times as great as that in air, which is about 1.3 times as great as that in the methane–air mixture in which the residual oxygen concentration is 19.2%. It is not probable that the slight difference of oxygen concentration, such as about 0.37%, results in the increase in the ignition delay time by 30%.

Figure 18 shows the comparison between the ignition delay times in 0.75% propane–air mixture and nitrogen–oxygen mixture with 18.49% oxygen concentration for the ambient temperature of 923 K. Although the residual oxygen concentration in propane–air mixture is predicted to be 17.25% at the elapsed time of 15 s, as shown in Fig. 8, the ignition delay time for propane–air mixture is smaller than that for the nitrogen–oxygen mixture. The results in Figs. 17 and 18 suggest that there exist some factors that promote the spontaneous ignition of droplets in fuel–air mixtures on the assumption that the prediction for the residual oxygen concentration is valid. The existence of radical species or intermediate species in the ambience formed by the chemical reaction can be considered to be one of the factors that can affect directly the reactions related to the ignition

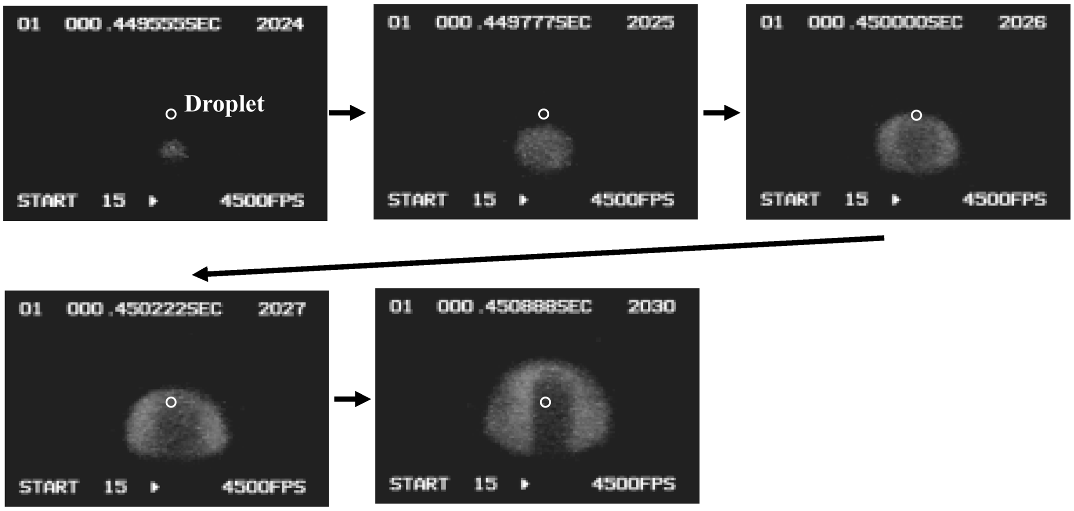

Fig. 19 OH band emission at spontaneous ignition of a droplet; frame speed is 4500 frames/s.

process. However, the promotion of spontaneous ignition of droplets by radical species in the ambience is not confirmed in this work because of some limitations on the experimental approach. One of the limitations is the difficulty in estimating precisely the residual oxygen concentration in the ambient gas. That is, the discrepancies in Figs. 17 and 18 may be attributable to the uncertainty of the residual oxygen concentration. There may be a possibility that the residual oxygen concentration in fuel–air mixture is larger than predictions. The existence of the temperature distribution in the electric furnace may also result in the nonuniform chemical reaction in the fuel–air mixture. A more sophisticated experimental setup and procedure should be used. In addition, the numerical simulation with detailed chemical reaction model[31] can be a useful tool for the understanding of the effect of the existence of radical species in the ambience on the droplet ignition.

It is well known that the ignition delay time for spontaneous droplet ignition is affected by the natural convection. Figure 19 shows the OH band emission image just after ignition recorded by the high-speed camera with an image intensifier. The first OH emission occurs in the region below the droplet and then it surrounds the droplet. This is because the evaporated fuel gas flows downward due to natural convection, which is similar to the results in previous studies.[11,16] However, microgravity experiments[16] have shown that the differences of the ignition delay time are quite small between normal gravity and microgravity, which suggests that the effect of natural convection on ignition delay time is small.

Conclusions

Experiments were carried out of the spontaneous ignition of n–dodecane droplets in lean methane–air, propane–air, and n–heptane–air mixtures under high-temperature environments, and the ignition delay time was measured. The variation of concentration of chemical composition with time in fuel–air mixtures was predicted by the zero-dimensional calculation of the chemical reaction with a detailed elementary reaction scheme, and the effects of change in chemical composition in the ambient mixture on droplet ignition process were discussed.

Experimental results showed that the ignition delay time in fuel–air mixtures is almost the same as that in air in the case of the lower ambient temperature, where the most of fuel and oxygen in the ambient mixture survive. This suggests that a small amount of fuel in the ambience has little effect on the droplet ignition process, which agrees with analytical results in a previous study. Note that this result can be obtained only in the case that the methane is added in the ambience. On the other hand, the ignition delay time in fuel–air mixtures becomes larger than that in air in the case of the higher ambient temperature. In this case, the prediction showed that most of fuel is consumed completely and the oxygen concentration decreases in the ambience by the chemical reaction. Based on these results, it can be concluded that one of the reasons why the ignition delay increases in fuel–air mixtures is the decrease in the oxygen concentration. However, the experimental result also shows that the ignition delay time in fuel–air mixtures is found to be longer than that in nitrogen–oxygen mixtures that have the same oxygen concentration as the residual one in fuel–air mixtures. That suggests that the change in the ignition delay time can not be only explained by the decrease in the oxygen concentration in the ambience by the chemical reaction. There is the uncertainty of the residual oxygen concentration in the fuel–air mixtures in experiments. The numerical simulation can be a useful tool for better understanding of the droplet ignition process in fuel–air mixtures.

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References

[1] Faeth, G. M., and Olson, D. R., "The Ignition of Hydrocarbon Fuel Droplets in Air," *SAE Transactions*, Vol. 77, 1968, pp. 1793–1802.

[2] Wood, B. J., and Rosser, W. A., Jr., "Experimental Study of Fuel Droplet Ignition," *AIAA Journal*, Vol. 7, No. 12, 1969, pp. 2288–2292.

[3] Sangiovanni, J. J., and Kesten, A. S., "A Theoretical and Experimental Investigation of the Ignition of Fuel Droplets," *Combustion Science and Technology*, Vol. 16, Nos. 1–2, 1977, pp. 59–70.

[4] Law, C. K., "Theory of Thermal Ignition in Fuel Droplet Burning," *Combustion and Flame*, Vol. 31, 1978, pp. 285–296.

[5] Moriue, O., Eigenbrod, C., Rath, H. J., Okai, K., Tsue, M., and Kono, M., "Effects of Dilution by Aromatic Hydrocarbons on Staged Ignition Behavior of n-Decane Droplets," *Proceedings of the Combustion Institute*, Vol. 28, 2000, pp. 969–975.

[6] Moriue, O., Eigenbrod, C., Rath, H. J., Tsue, M., Kono, M., and Sato, J., "Spontaneous Ignition of n-Alkane Droplets with Various Volatility," *Journal of the Japan Society for Aeronautical and Space Sciences*, Vol. 47, No. 157, 2004, pp. 189–194.

[7] Takei, M., Tsukamoto, T., and Niioka, T., "Ignition of Blended-Fuel Droplet in High-Temperature Atmosphere," *Combustion and Flame*, Vol. 93, Nos. 1–2, 1993, pp. 149–156.

[8] Tsukamoto, T., Odaka, H., and Niioka, T., "Ignition Simulation of a Fuel Droplet in High-Pressure High-Temperature Atmosphere," *Transaction of Japan Society for Aeronautical and Space Sciences*, Vol. 35, No. 110, 1993, pp. 165–176.

[9] Kadota, T., Satoh, K., Segawa, D., Sato, J., and Marutani, Y., "Autoignition and Combustion of a Fuel Droplet in Supercritical Gaseous Environments Under Microgravity," *Proceedings of the Combustion Institute*, Vol. 27, 1998, pp. 2595–2601.

[10] Segawa, D., Kadota, T., Nakainkyo, A., Hirota, S., and Enomoto, H., "Effects of Ambient Pressure on Autoignition of a Fuel Droplet in Supercritical and Microgravity Environment," *Proceedings of the Combustion Institute*, Vol. 28, 2000, pp. 1063–1069.

[11] Nakanishi, R., Kobayashi, H., Kato, S., and Niioka, T., "Ignition Experiment of a Fuel Droplet in High-Pressure High-Temperature Ambient," *Proceedings of the Combustion Institute*, Vol. 25, 1994, pp. 447–453.

[12] Huang, L.-U., and Chen, C.-H., "Droplet Ignition in a High-Temperature Convective Environment," *Combustion and Flame*, Vol. 109, Nos. 1–2, 1997, pp. 145–162.

[13] Niioka, T., Kobayashi, H., and Mito, D., "Ignition Experiment on Droplet Array in Normal and Microgravity Environments," *IUTAM Symposium on Mechanics of Droplets and Arrays*, 1994, pp. 367–377.

[14] Goto, M., Ju, Y., and Niioka, T., "Numerical Analysis of Ignition of Fuel Droplet Array in Hot Stagnant Air," *Proceedings of the Combustion Institute*, Vol. 27, 1988, pp. 1959–1966.

[15] Kadowaki, T., Maruta, K., Hasegawa, S., Kobayashi, H., and Niioka, T., "Microgravity Experiments on Ignition of Droplet Matrix," *Journal of the Japan Society of Microgravity Application*, Vol. 14, No. 1, 1997, pp. 17–24 (in Japanese).

[16] Tanabe, M., Kono, M., Sato, J., Koenig, J., Eigenbrod, C., and Rath, H. J., "Effect of Natural Convection on Two Stage Ignition of an n-Dodecane Droplet," *Proceedings of the Combustion Institute*, Vol. 25, 1994, pp. 455–461.

[17] Tanabe, M., Bolik, T., Eigenbrod, C., Rath, H. J., Sato, J., and Kono, M., "Spontaneous Ignition of Liquid Droplets from a View Point of Non-Homogeneous Mixture Formation and Transient Chemical Reactions," *Proceedings of the Combustion Institute*, Vol. 26, 1996, pp. 1637–1643.

[18] Schnaubelt, S., Moriue, O., Coordes, T., Eigenbrod, C., and Rath, H. J., "Detailed Numerical Simulations of the Multistage Self-Ignition Process of n-Heptane Isolated Droplets and Their Verification by Comparison with Microgravity Experiments," *Proceedings of the Combustion Institute*, Vol. 28, 2000, pp. 953–960.

[19] Yang, J.-R., and Wong, S.-C., "On the Suppression of Negative Temperature Coefficient (NTC) in Autoignition of n-Heptane Droplets," *Combustion and Flame*, Vol. 132, No. 3, 2003, pp. 475–491.

[20] Shaygan, N., and Prakash, S., "Droplet Ignition and Combustion Including Liquid-Phase Heating," *Combustion and Flame*, Vol. 102, Nos. 1–2, 1995, pp. 1–10.

[21] Segawa, D., Kadota, T., Kohama, R., and Enomoto, H., "Ignition of Binary Mixture Droplets by a Propagating Laminar Flame," *Proceedings of the Combustion Institute*, Vol. 28, 2000, pp. 961–968.

[22] Niioka, T., Ishiguro, S., and Saito, T., "A Numerical Approach to Fuel Droplet Ignition," National Aerospace Lab., Technical Rept. TR-628T, 1980.

[23] Kaskan, W. E., "The Dependence of Flame Temperature on Mass Burning Velocity," *Proceedings of the Combustion Institute*, Vol. 6, 1957, pp. 134–143.

[24] Westbrook, C. K., and Dryer, F. L., "Chemical Kinetics and Modeling of Combustion Process," *Proceedings of the Combustion Institute*, Vol. 18, 1981, pp. 749–767.

[25] Warnatz, J., Maas, U., and Dibble, R. W., *Combustion*, 3rd ed., Springer-Verlag, Berlin, 2000, pp. 119–122.

[26] Zabetakis, M. G., Lambiris, S., and Scott, G. S., "Flame Temperatures of Limit Mixtures," *Proceedings of the Combustion Institute*, Vol. 7, 1959, pp. 484–487.

[27] Koert, D., Pitz, W. J., Bozzelli, J. W., and Cernansky, N. P., "Chemical Kinetic Modeling of High Pressure Propane Oxidation and Comparison to Experimental Results," *Proceedings of the Combustion Institute*, Vol. 26, 1996, pp. 633–640.

[28] Gear, C. W., *The Automatic Integration of Stiff Ordinary Differential Equations, Information of Processing*, edited by A. J. H. Morrell, North-Holland, Amsterdam, 1969, pp. 187–193.

[29] Pitsch, H., Peters, N., and Seshadri, K., "Numerical and Asymptotic Studies of the Structure of Premixed iso-Octane Flames," *Proceedings of the Combustion Institute*, Vol. 26, 1996, pp. 763–771.

[30] Biet, J., Delfau, J.-L., Seydi, A., and Vovelle, C., "Experimental and Modeling Study of Lean Premixed Atmospheric-pressure Propane/O_2/N_2 Flames," *Combustion and Flame*, Vol. 142, No. 3, 2005, pp. 197–209.

[31] Marchese, A. J., Dryer, F. L., and Nayagam, V., "Numerical Modeling of Isolated n-Alkane Droplet Flames: Initial Comparison with Ground and Space-based Microgravity Experiments," *Combustion and Flame*, Vol. 116, No. 3, 1999, pp. 432–459.