Ignition of Hydrogen-Oxygen Premixed Gases with Excimer Laser

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ABSTRACT

Recently photochemically augmented combustion techniques have been researched in order to shorten ignition delay and to accelerate the burning velocity of reactive gases by irradiation from ultraviolet light sources. However, the phenomena of the ignition and the flame growth still have not been observed thoroughly and the photochemical reactions have yet to be clarified.

At first step, authors have carried out experiments on the ignition for hydrogen-oxygen-argon mixtures by using an ArF excimer laser to observe the phenomena of ignition and flame growth by using a high speed camera and the shadowgraphy, and also to evaluate the minimum incident laser energy for the ignition under various mixture conditions.

A series of shadowgraphs indicated that the ignition occurred in the laser focal region and the initial flame kernel created by the laser ignition grew suddenly along the optical path. This kernel growth was different from the kernel growth due to laser break-down. It then grew with two projections at both sides of focal point in laser axis direction. The flame growth speed was greater in the laser axis direction than in the perpendicular direction. The difference of the propagating speeds between the direction of laser axis and the perpendicular direction increased with the laser energy increasing. Therefore, the ignition and the initial flame growth were augmented by the irradiation of the laser light. The minimum incident laser energy was found to be related to oxygen partial pressure within the experimental ranges. When the laser optical path was filled with nitrogen to avoid the absorption by oxygen molecules in the air, the minimum incident laser energy was about 32 % lower in average than when filled with air. It was therefore confirmed that the photodissociation of oxygen molecules into oxygen atoms plays an important role in the ignition process.

INTRODUCTION

Clean combustion and high thermal efficiency have been always key words in combustion applications since the environmental problems of the 1970's. To attain these targets, much new combustion technology has been researched and developed. However, combustion technology has been still limited by the combustion characteristics of reactive mixtures such as flammability limit, and burning velocity. To take away these constraints physically and/or chemically, various augmented combustion techniques have been studied by use of plasma jet(1), photolysis(2), electric or magnetic fields, and catalysts. Recently much attention has been given to flash photolysis(3) which augments combustion photochemically.

M. Lavid and coworkers(4) researched theoretically and experimentally the ignition of hydrogen-oxygen and hydrogen-air mixtures with excimer laser. The photochemical ignition was achieved by using the F_2 laser with 157 nm of center wavelength which is in a strong absorption band for oxygen molecules. When using the ArF laser with 193 nm which is in a weak absorption band near the edge of the O_2 Shumann-Runge band, although the calculated minimum light fluence was very high 2.7×10^5 mJ/cm^2, the ignition occurred actually around 10^3 mJ/cm^2. They predicted that the ignition due to the ArF laser was probably caused by another mechanism different from single photon photochemistry of O_2 into O atoms.

B. E. Forch and coworkers(5) studied the ignition properties of premixed hydrogen-oxygen flow by using the tunable laser system, and indicated that the three minima in incident laser energy for the ignition appeared on the three wavelengths, which exactly corresponded to the two-photon resonant wavelengths of oxygen atoms(6). They concluded that the ignition phenomena occurred by the three steps of (1) multiphoton photochemical formation of oxygen atoms, (2) multiphoton ionization of these atoms, and (3) the formation of laser microplasma using the electrons formed in the previous process.

This combustion technique, by using photochemical reaction, has potential to extend the limits of the combustion properties and also to control the chemical process of the combustion directly as an in-situ technique in the near future. However, the phenomena of these ignitions and accelerations of flame propagation by irradiation of ultraviolet lights have not still been observed thoroughly enough and also the reaction mechanism including photochemical reaction has not been made clear enough for this new combustion technology to be realized.

In this first paper, the authors carried out experiments on ignition for hydrogen-oxygen-argon mixtures by use of ArF excimer laser using the 300mJ high power as an ultraviolet light source, and observed the phenomena of the ignition and the flame growth by using a high speed camera and the shadowgraphy. We also evaluated the minimum incident laser energy needed to ignite mixtures under various oxygen concentrations and the initial pressure of mixtures.

EXPERIMENTAL APPARATUS

The outline of the ignition chamber is shown in Figure 1. The combustion section has three cylindrical holes each with a diameter of 40 mm, and the center axis of the each hole is perpendicular to the axis of the other holes and crosses center point of the ignition chamber. An optical grade quartz window is set up at both ends of the two cylindrical holes to be used as an optical path for ignition laser and for shadowgraphy. At both sides of the third hole, a mixture inlet, a burned gas outlet, and the measuring ports for temperature and pressure are arranged. The chamber has the focus lens of f=100 mm, which is attached just before the window for the excimer laser, so that the laser is focused at the center of the chamber.

The schematic diagram of the experimental apparatus is shown in Figure 2. The ArF laser with a maximum energy of

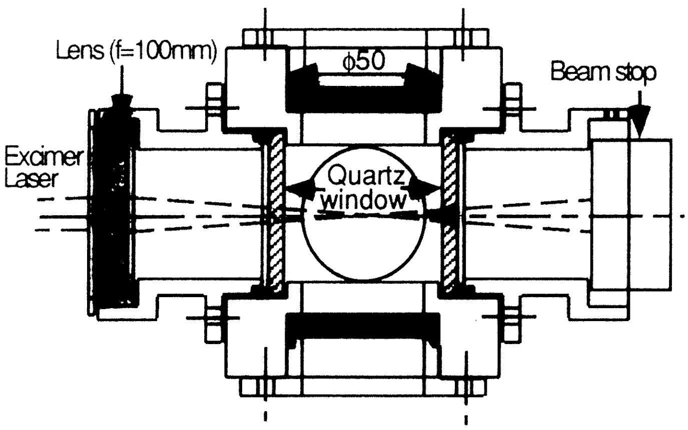

Fig.1 Ignition chamber

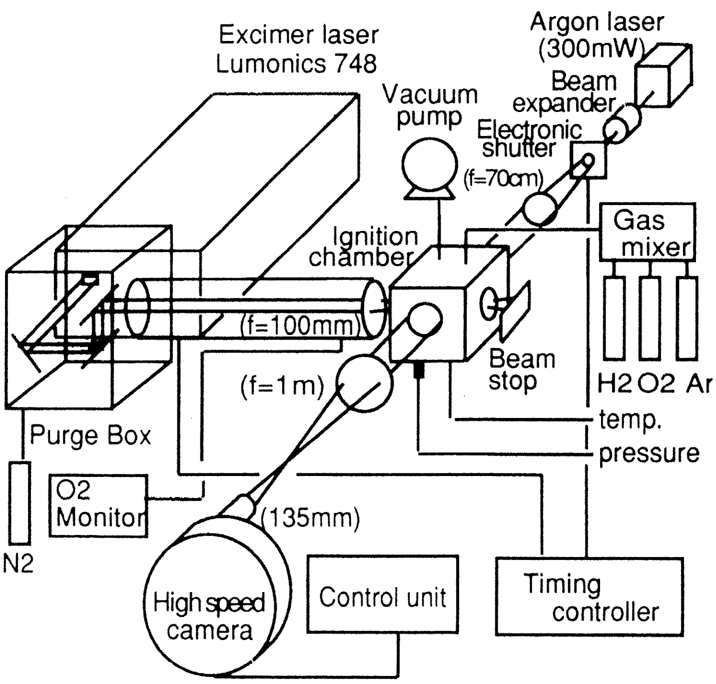

Fig.2 Experimental apparatus

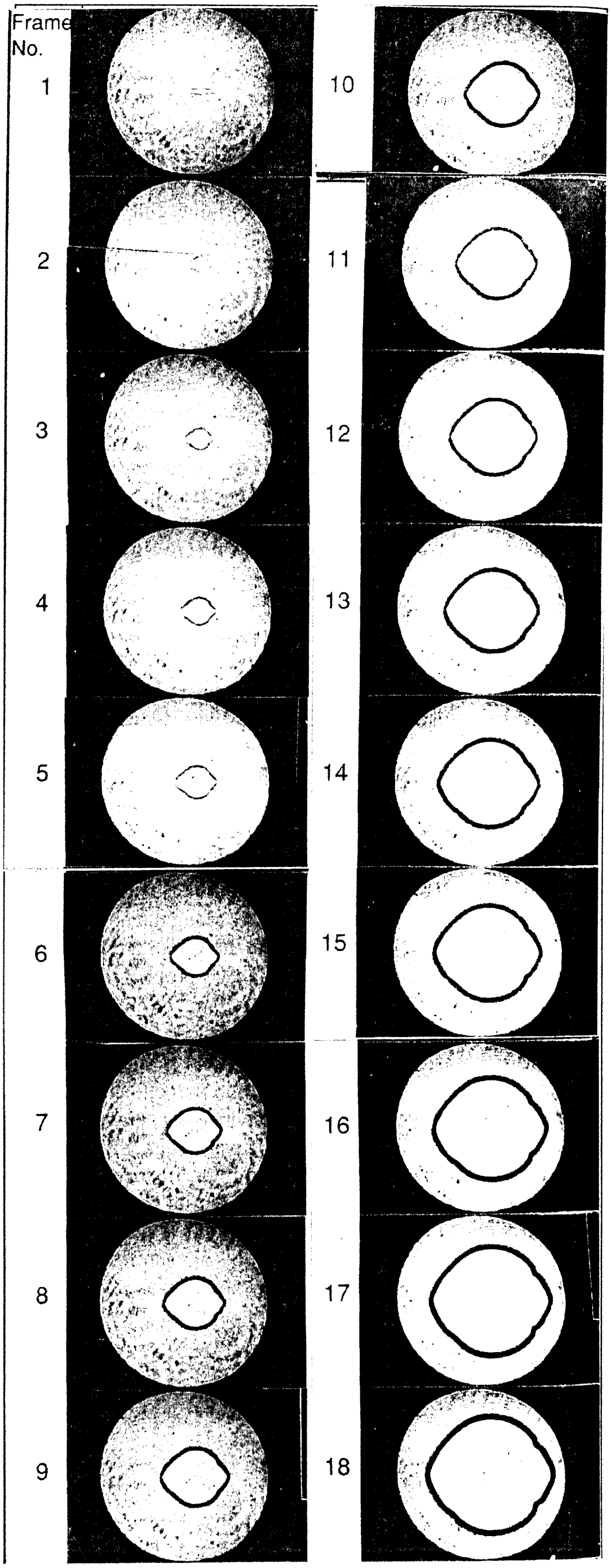

Fig.3 Picture of growing flame

H2:20%,O2:10%,Ar:70%(ϕ =1.0) Initial gas pressure:0.08MPa
Frame speed:10000frames/s Laser power 286mJ
Optical path filled with N2

300 mJ (Lumonics 748) is used as an ultraviolet light source. The laser beam with 9 mm x 23 mm rectangular section is reflected by three mirrors on which the direction and the height of the beam are adjusted, and focused at the focal point in the ignition chamber. A purge box filled with nitrogen is attached along the optical path from the laser port to the focus lens to prevent light being absorbed by the oxygen in the air. A laser shadowgraph system and a high speed drum camera (Cordin 350) are set up, so that the ignition and flame growth can be observed.

This experiment is carried out by using hydrogen-oxygen-argon premixed gases with various initial pressures below atmospheric pressure and various oxygen concentrations, when the optical path from the laser outlet to the focus lens is filled with nitrogen or with atmospheric air.

EXPERIMENTAL RESULTS

Observation of Ignition and Flame Growth

As has already been mentioned, the Lavid's paper points out the large differences of incident laser energy between the calculated values and the experimental values when the ArF laser is used as an ultraviolet light source. First of all, the phenomena for the ignition process was observed by irradiation of ArF excimer laser.

Figure 3 shows typical shadowgraphs for the ignition and flame propagation. In this case, the premixture had the concentration of 20% H_2, 10% O_2, and 70% Ar and initial gas pressure of 0.08 MPa. Incident laser energy was 286 mJ and the optical path of the excimer laser was filled with N_2. The camera frame speed was 10000 frame/sec which corresponded to the exposure time 2.7 ms. The laser beam came from the left side of each photo frame and went through to the right of the pictures.

A series of photographs indicate that two straight streaks appeared firstly in the focal region of the laser immediately after the laser irradiation, and in the same first frame. The two streaks shows the schliere of up and down peripheries of a horizontal volume due to the variation of the refractive index of the mixture. The flame kernel occurred suddenly at the two streaks. After that, the flame kernel grew, keeping an elliptical shape with projections in the direction of laser optical axis, which looked like a lemon. The projection in the laser incident side was bigger than in the other side.

The phenomena for the ignition and the initial flame growth were compared as incident laser energy was changed until the ignition no longer occurred. These photographs are shown in Figure 4. The experimental conditions were the same as in the case of Figure 3 except for incident laser energy. With incident laser energy of 169 mJ as shown in case 5 of Figure 4, ignition did not occur.

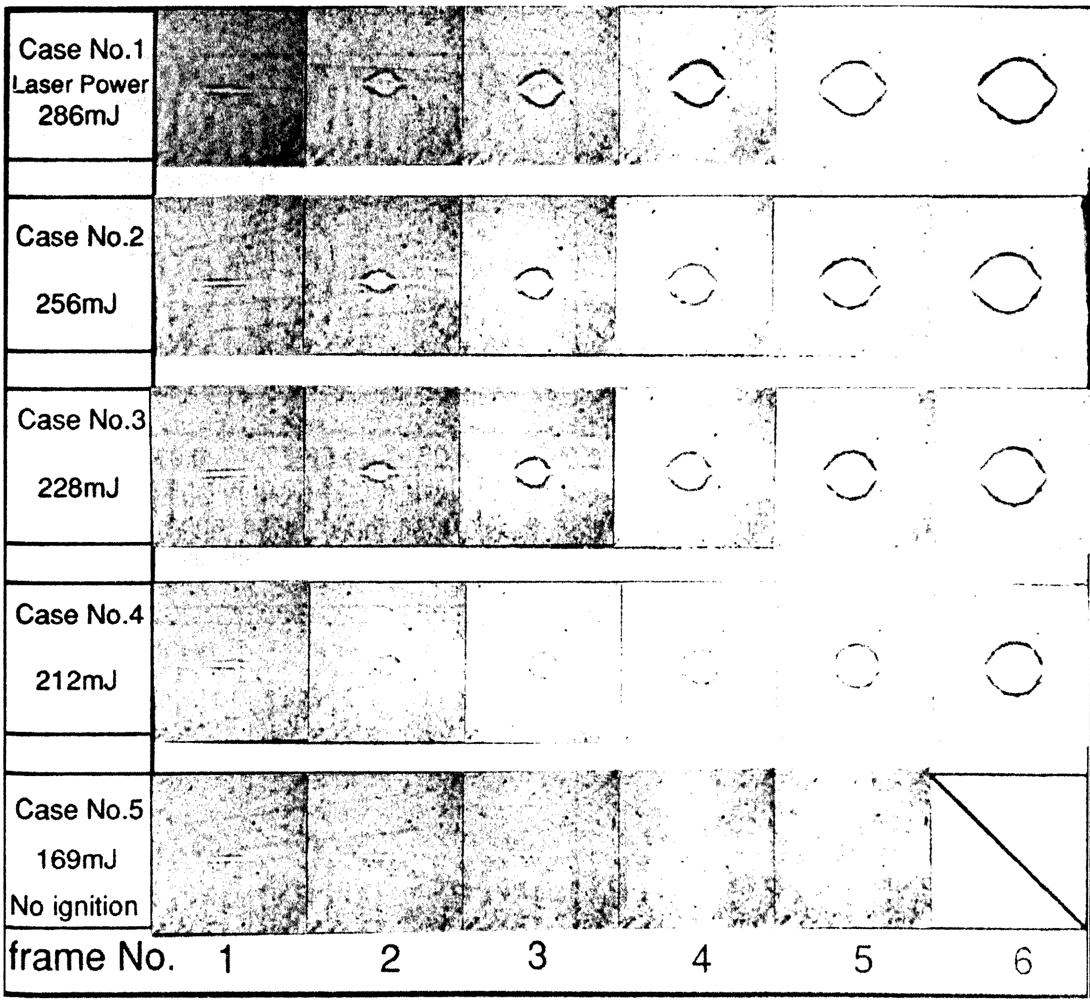

Fig.4 Picture of ignition and flame growth on each laser power
(H2:20%,O2:10%,Ar:70% (ϕ =1.0) Initial gas pressure:0.08MPa
Frame speed:10000frames/s Optical path filled with N2)

The two straight streaks due to the laser irradiation were observed in the first frame under the condition of each incident laser energy. A series of photos in case 5 indicate clearly that these streaks decreased as the time passed, although they existed 2 or 3 frames corresponding to 2~300 ms. As the ignition occurred the center of these streaks and flame propagation started from high contrast part of the streaks irrespective of laser energies. The projections of flame were higher as the incident laser energy was higher. This flame kernel did not have characteristic shape which has some explosive kernels due to laser breakdown.

Flame Growth Speed

To grasp the effect of ArF laser energy to the initial flame growth quantitatively, the authors measured the diameters of the flame in the laser direction (Dh) and in the perpendicular direction (Dv) from the shadowgraph pictures. As shown in Figure 5, the results explain that the diameter Dh of the flame kernel, particularly in laser direction, enlarged rapidly during 2~300 ms. After that, the diameter enlarged linearly with time.

Therefore, these flame growth speeds V deviated from the linear approximation for the each data except the data within 2~300 ms in figure 6. As the results show, the propagating speed Vh in the laser direction was higher than the speed Vv in the perpendicular direction. At the early stage of the flame propagation, the effect of accelerating the speed was greater as the incident laser energy was higher.

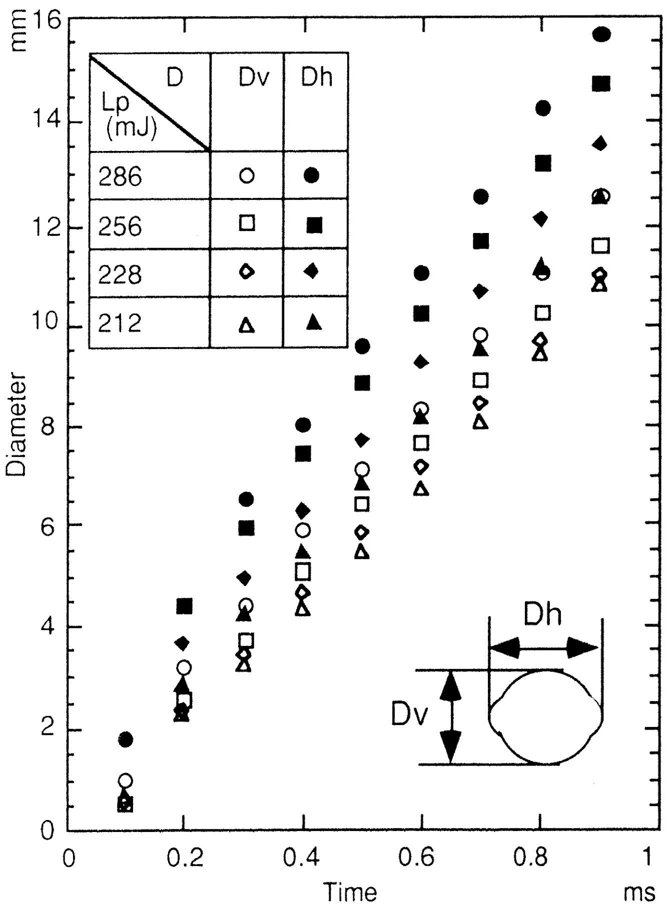

Fig.5 Comparison of flame growth in the direction of laser path with in the perpendicular

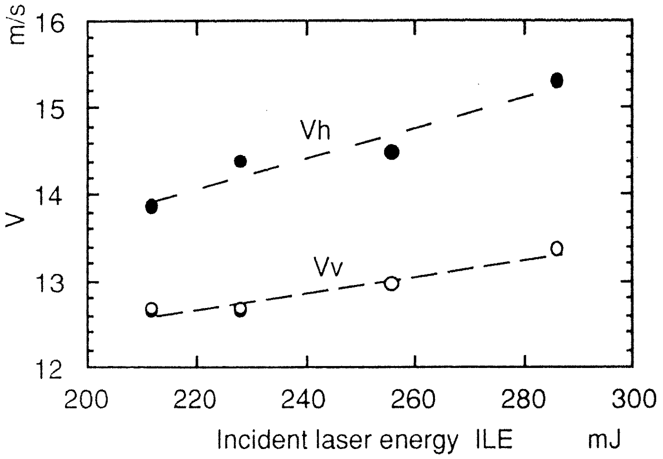

Fig.6 Effect of incident laser energy to initial flame propagating speed

Minimum Incident Laser Energy

The minimum incident laser energy ILE needed to ignite mixtures was evaluated by changing the oxygen concentration and the initial pressure of premixtures under the constant hydrogen concentration of 20%. In this experiment, the effect of oxygen in the optical path from the laser port to the focus lens was also examined. The results are shown in Figure 7(a),(b), where (a) is the case of the laser optical path purged by dry N_2 gas and (b) the case of the path without the purge. In these figures, minimum incident laser energy is the relative value to the incident laser energy, when O_2 concentration is 10% at initial pressure of 0.08 MPa and the optical path without the N_2 purge. In these experimental conditions, minimum incident laser energy was lower as the initial gas pressures and/or oxygen concentrations were higher. The effects of these factors were almost same qualitatively between both optical paths. The phenomena for the ignition and the flame propagation also were observed to have no difference of both gases in the laser optical path.

Generally, the light energy of excimer laser with ultraviolet wavelength decreases due to the effect of the atmospheric air in optical path, since oxygen molecules in the air have absorption bands beyond 250 nm[8]. We estimated the power spectrum profile for the irradiation system of the ArF laser by using the spectral absorption coefficient of oxygen molecule [9], when it

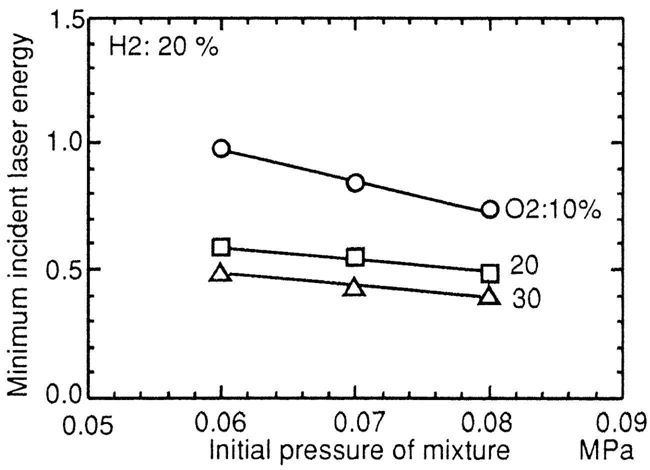

Fig.7 (a) Minimum incident laser energy under optical path filled with Air

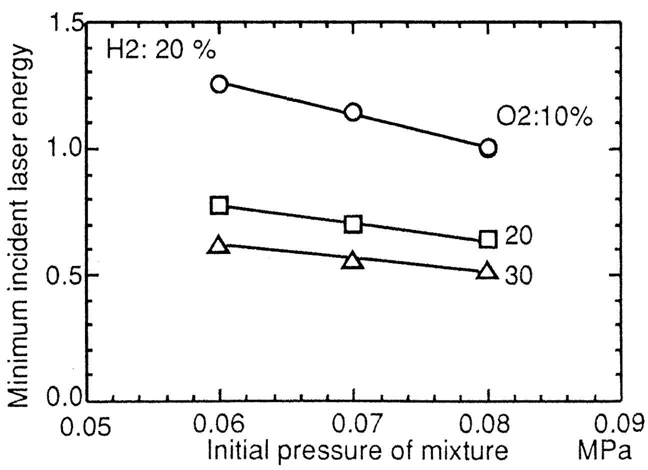

Fig.7 (b) Minimum incident laser energy under optical path filled with Air passed through the optical path which was purged by N_2 or not as shown in Figure 8. In the case of the purged optical path, the laser passed through air for the distance of 15 cm, which is the unavoidable optical path inside the laser system. In this case, the incident laser energy in each of the wavelengths absorbed little by O_2. When the optical path was not purged, each power with the wavelengths absorbed by O_2 in the air decayed as the total distance filled with air was 165 cm. In comparison with (a) and (b) in Figure 7, minimum incident laser energy in the purged case was actually about 32 % lower in average than the unpurged case. Thus, in our experiments the power spectrum profile and total power were predicted to be effected by O_2 in the optical path of the ArF laser.

As it was predicted the only absorption molecule is O_2 in this experiment, we replotted the ILE data versus the partial pressure of oxygen. This is shown in Figure 9. It was found that

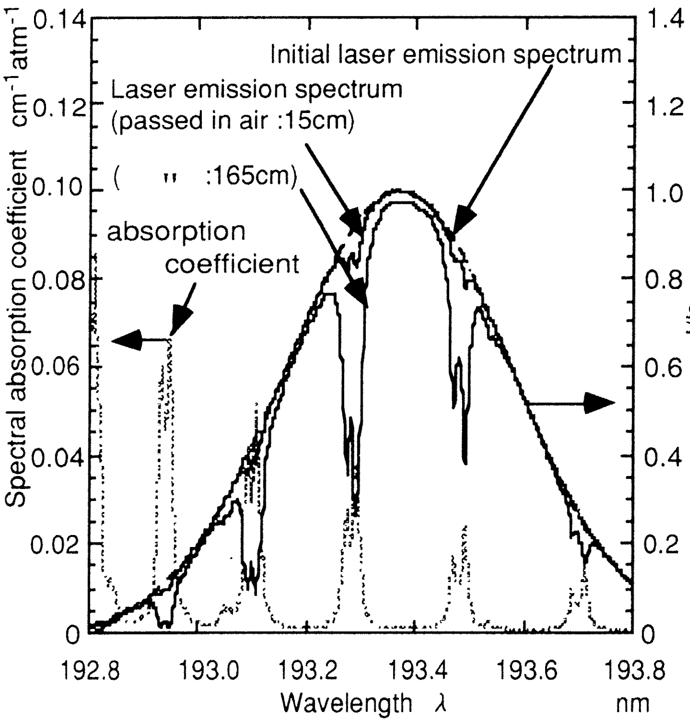

Fig.8 Effect of the optical path to the laser incident energy spectrum

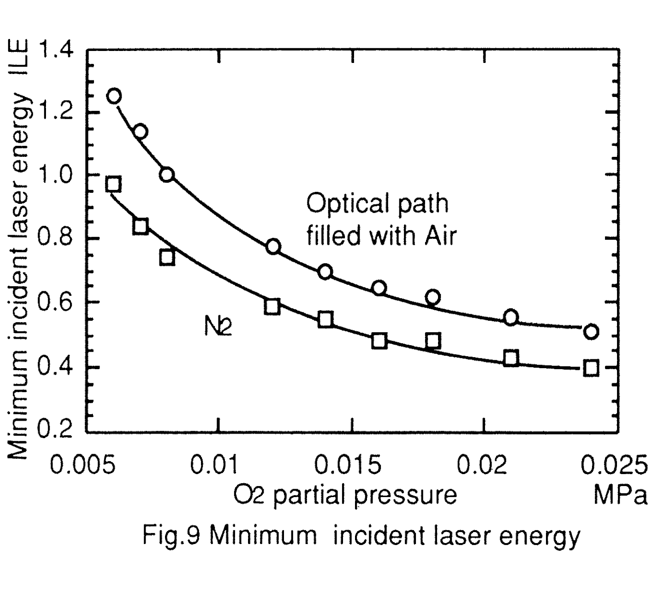

Fig.9 Minimum incident laser energy

all ILE data was expressed as some function of oxygen partial pressure, in which the minimum incident laser energy decreased with the increase of oxygen partial pressure.

DISCUSSION

Possibility of the Break-down Ignition

A focused high power laser is known to bring break-down and make a plasma. If the break-down occurs, the laser fluence changes drastically before and after the focal point, moreover the laser fluence through the focal point particularly does not increase even if incident laser energy is increased. The authors checked with a power meter that there was no difference of laser fluence before and after the focal point, and that the fluence after the focal point was not saturated. Furthermore, no special noise due to the break-down could be heard by irradiation of the laser.

The maximum fluence of excimer laser at the focal point was estimated about 10^{10} W/cm^2, which was calculated from the width between the two streaks near the focal point on the shadowgraph. This fluence was lower than the minimum break-down fluence 10^{13} W/cm^2 of Nd-YAG laser in argon[(10)], which was lower than the fluence of excimer laser.

Moreover, the flame kernel was observed to be different from that due to break-down[(7)], and the dependence of oxygen partial pressure on the minimum incident energy was recognized as mentioned above.

Therefore, all these items discussed above explained that the possibility of the break-down ignition was quite low.

Two Straight Streaks on the Laser Path

Two straight streaks in the every first frame of shadowgraph just after laser oscillation, indicate the variation of the refractive index of the mixture. This may be caused by the change of gas composition or temperature in these streaks. In this experiment, the amount of oxygen atoms generated by irradiation of laser, was estimated to be almost 10^7 mol/cm^3. Then the change of gas composition was too small to be the cause of this difference. On the other side, the quenching reactions of O atoms and hydrogen-oxygen chain reactions can be produce the heat release. The authors estimated the orders of the temperature rise and reaction rate for each elemental reaction in the conditions which were oxygen concentration of 10%, the incident laser energy of 200 mJ, initial gas pressure of 0.08 MPa, initial gas temperature of 300K, and in the optical path purged by N_2 using the absorption coefficient in Figure 8. Then, if all photochemically dissociated O atoms recombined into O_2 molecules by the quenching reactions only, the mixture temperature was increased by about 30K at the cross section based on the width of the streaks and 200K at the ideal laser focal cross section. Temperature rise does not deny the occurrence of the streak. In the consumed reactions of O atoms, however, O atom is consumed not only by the quenching reaction of O atoms directly into O_2 molecule, but also by O_3 formation reaction. Therefore, at the next step, it is necessary to carry out the numerical analysis for the reactions including photochemical reaction and observation of the ignition process in more detail using the ultra-high speed camera.

Ignition Mechanism

If the ignition phenomena in these experiments were mainly controlled by the photochemical reaction O_2+hv=O+O, such as single-photon photochemistry, the production amount of oxygen atoms is proportionate to the product of oxygen partial pressure P_{O2} by minimum incident laser energy ILE. Then if the ignition occurs when the concentration of oxygen atoms reaches a constant value of P_{O2} x ILE=constant, ILE is inversely proportional to oxygen partial pressure. On the basis of this view, all data in Figure 9 was replotted versus the reciprocal of oxygen partial pressure. As the results show in Figure 10, the data were on a straight line, although minimum incident laser energy is not inversely proportional to of oxygen partial pressure.

Moreover, when the laser fluence for the ignition is compared between this experimental data and the calculated data according to single-photon photochemistry(4), the experimental laser fluence was same order as the calculated one.

Therefore, it is predicted that the amount of produced oxygen atoms played an important role in this ignition phenomena. However, we could not conclude that the ignition was caused by single photon photochemistry within this experiment ranges, since we can also consider this ignition occurred by the microplasma according to multiphoton photochemistry(5).

In this work, the authors were able to observe the interesting phenomena for the ignition and initial flame growth by ArF excimer laser. At the same time, we also found many questions and obscure points. To solve them, we plan to research this photochemical combustion with spectrochemical analysis and the numerical analysis of chemical reactions.

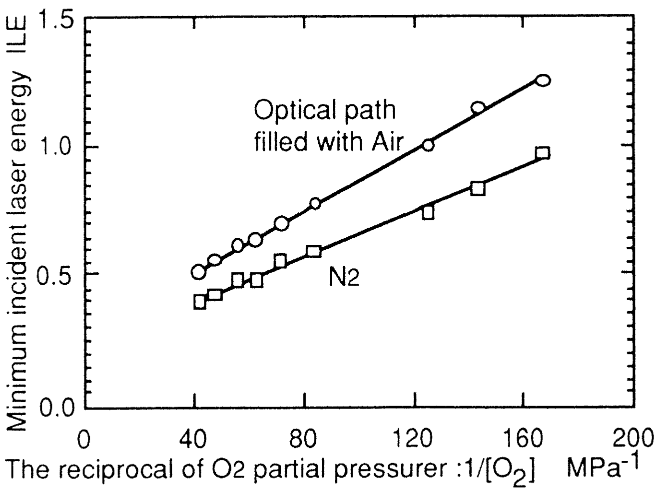

Fig.10 Minimum incident laser energy

CONCLUSION

The results on the ignition and the flame propagation of hydrogen-oxygen-argon premixed gases by irradiation of the ArF excimer laser reached following conclusions.

(1) The ignition occurred on two streaks in the focal area just after the laser oscillations. The initial flame kernel firstly grew suddenly on the streaks within a few hundred microseconds and then propagated keeping an elliptical shape with two projections in laser optical direction. These phenomena of the ignition are different from the one due to break-down.

(2) The propagating speed in the direction of the laser optical path is higher than the perpendicular direction. The effect of augmenting the speed was greater when incident laser energy was higher.

(3) The minimum ignition laser energy is related linearly to the reciprocal number of oxygen partial pressure. It confirmed that oxygen atoms produced by photochemical reaction play a very important role in the ignition phenomena.

(4) The minimum ignition laser energy in the optical path purged by nitrogen is about 32 % lower in average than in the unpurged one. However, the difference of phenomena for the ignition process could not be observed and also the effects of the oxygen concentration and the initial pressure of mixtures to the laser energy were not changed qualitatively.

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