Curiosity of Shock-Compression Low-Temperature Ignition

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Abstract

We have examined and reasoned a curious phenomenon that the activation energy for the compression ignition of a methane mixture is reduced suddenly to the almost half when the temperature behind a reflected shock wave decreases down to a critical temperature around 1 600 K and ignition delay becomes longer than 1 ms. A mixing zone could be raised ahead of a contact surface probably at a rupture of a partition diaphragm. The mixing zone interacts with the reflected shock wave, and a rarefied wave is initiated. When an ignition delay is longer than 1 ms, the rarefied wave propagates through the mixture in which preflame reactions are in progress. The reactions would be accelerated and then a pressure rise will be caused under the effect of the rarefied wave. A steep pressure rise of real ignition starts near the end of the tube and propagates toward the driver section, but the ignition has been originated not at the end of the tube but a little to the inside of the tube. The ignition delays in lower temperature than the critical point are not the pure chemical delays under the pure conditions of constant temperature and pressure, given from the speeds of incident shock wave.

Introduction

A shock tube is expected to realize low-temperature compression ignition of mixtures without a long compression heat-up duration in the blue-flame dominant temperature regime [1]. Rapid-compression machines have been used for the compression devices, where the heat-up duration becomes relatively long, comparable to the ignition delays. This is the reason shock tubes should be introduced for the mixture compression. As the first step we have measured ignition delay of methane/oxygen/argon mixtures using a shock tube. The activation energy was reduced suddenly from 182 to 92 kJ/mol when the temperature behind a reflected shock wave decreased down to 1 600 K and after; at this critical temperature the ignition delay was about 1 ms. A behavior of this kind has been shown in references [2, 3, 4, 5]. If this change of activation energy is due to the change of reaction mechanisms depending on the temperature, we could find the way for shock tubes as a media for investigating low-temperature ignition, but if it is caused by other reasons depending on the apparatus itself, effect of fluid flow for example, ignition-delay measurements using shock tubes are meaningless. In this paper, this curious sharp decline of activation energy will be made clear, and it will be mentioned that a rarefied wave caused by an interaction between a reflected shock wave and a contact surface would be closely related to the trigger of ignition onset and that the position on the low-temperature ignition in shock tubes.

Experimentals

The shock tube used here was a tube, with an inner diameter 52.5 mm, a driver section length 2 200 mm, and a driven section length 3 300 mm. The pressure transducers and quartz windows were equipped in six positions along with the tube; 17, 178, 340, 503 665 and 990 mm distant from the end of the driven section, and each position is called herewith the observing position A, B, C, D, E and F. On the wall at each observing position three holes were arranged in zero, three and nine o'clock geometry. Each zero o'clock hole was fitted with a Kistler 601A pressure transducer, and three and nine o'clock holes, facing each other, were fitted with quartz windows. The test gas and mixture to be compressed were pure argon and a stoichiometric methane/oxygen/argon mixture with 89.5 percent argon content by volume. The driver gas was pure helium, or ethanol-vapor-contained helium. The initial pressure in the driven section was 70 mmHg in every case.

The temperature behind reflected shock wave was determined by a velocity based on the pressure response difference between positions A and C, taking into account of temperature dependence of heat capacity of the gases. Reflected shock waves and contact surfaces were set in the "Under-Tailored" condition in the experiments.

Arrival of contact surface to each position was detected by incident laser-light attenuation. Behind the contact surface, temperature is lower associated with higher density than that behind the incident shock wave. A density change will result in a transmitted light variation. The ethanol-vapor contained in driver gas would condense into liquid droplets when the temperature falls, which would give us an distinct incident laser-light attenuation. Only by the density increase of the gas, the incident laser-light attenuation can be obtained. Light sources of solid-state laser (Matsushita LN9705, 5 mW, 785 nm in wave length) arranged to be parallel beams through collimator lens, and were introduced into the tube. Attenuated but remaining transmitted light was detected by photodiodes (Hamamatsu Photonics, S1336-18BQ, 190~1 100 nm).

Results and Discussion

Detection of contact surface

It will be mentioned first that the measurement of attenuated level of incident light intersected perpendicularly to the tube is effective to detect the arrival of contact surface; observing the transmitted laser-light level attenuated by schlieren effect raised in contact surface. Experimental results in a case the compressed gas in driven section is pure argon will be shown. Transmitted light levels at the positions C, E and F were recorded, and pressure was sampled at positions A, B, C and F. The temperature behind reflected shock wave was 1 479 K. Figure 1 shows a pressure trace of position A. Incident laser-light attenuation and pressure histories of position E are shown in Fig. 2, where the transmitted light swings upwards as the incident light is attenuated.

It can be seen from the pressure trace of Fig. 2 that the time I_5 is an arrival of incident shock. On the light trace after the arrival of incident wave, the transmitted light starts to decrease from the time M_5 and then declines steep at the time C_5. A similar history can be obtained at position F. At the position C such a steep attenuation cannot be observed as the one at C_5 of position E. At the position A, the most close position to the end of the tube, the pressure behind the reflected shock wave falls slightly at the time E_1 and then furthermore at the time of K_1 as shown in Fig. 1. A similar history can be obtained at position B. At the position C such a slight pressure fall cannot be observed as the one at E_1 of position A.

Position and time characteristic behaviors of pressure and transmitted-light histories can be well recognized when they are shown in a time-to-distance diagram. In Fig. 3 the present experiment is drawn. The abscissa is the distance from the partition diaphragm membrane and the ordinate the elapsed time after the incident shock wave has arrived at the end of driven section. Responses in each position are connected by solid straight lines and the extrapolated responses are shown by broken lines. Because of the under-tailored interaction condition between reflected shock wave and contact surface, a rarefied region; gas of low density, propagates at the sonic velocity behind reflected shock wave after the interaction between the reflected shock wave and contact surface. A temporary line starting from the time K_1 of Fig. 1 can be assumed in Fig. 3 with a gradient of sonic velocity therein, and the timings K_1, K_2 and K_3 sit on the line as straight as a single line. It can be seen from this fact that the timings K_1, K_2 and K_3 are the arrivals of the identical rarefied region. We tied together the timings C_5 and C_6 of steep attenuation of incident laser light at the positions E and F respectively, and drew the line out, then an intersection point α has been fixed on a line of reflected shock wave; R_3-R_5. Another intersection point β was fixed on the same line by adopting the extrapolated timing line of rarefied region K_1-K_3, and both points α and β were not far off.

The gradient of segment C_5-C_6, a propagating velocity of a phenomenon of steep incident-light attenuation is 533 m/s, which is very close to the one of moving velocity of contact surface, 492 m/s calculated from the incident shock-wave velocity. It is perceived that at the moment the incident-light attenuated the driver gas has arrived to the observed position. A situation is conceivable that a contact surface would go along the segment C_6-C_5, then interacts with the reflected shock wave, which would result in a formation of rarefied region propagating to the end of the tube along the segment K_3-K_1. It will be a good ground that the observation resembles the calculation in propagating velocity.

As recognized from the trace at the time of E_1 in Fig. 1, the pressure falls slightly before the rarefied region comes up to the position A. Here the rarefied region could be caused by an interaction between a contact surface and a reflected wave. In position E, the transmitted light also starts to decline behind the incident shock wave from the time M_5 as shown in Fig. 2. We will try to see the relation between the slight pressure drop of E_1 and the light attenuation at the time M_5. The pressure-drop moments E_1 and E_2 in positions A and B when tied together will give us a velocity of 689 m/s, which is very close to the sonic speed 716 m/s in that place. The moments transmitted light starts to decline, M_6, M_5 and M_3, can be linked together by almost a single line. An intersection point of extrapolated line of segments M_3-M_5 and the R_2-R_3 line of reflected wave is located in the vicinity of the intersection point of extrapolated line of segments E_1-E_2 and the R_2-R_3 line.

An incident shock wave is formed herewith by a rupture of a partition diaphragm. Therefore, a mixing zone could be raised ahead of a contact surface, wherein driver and driven gases are mingled [6]. As the interacting condition in these experiments carried out here is under tailored, the rarefied region will propagates behind the reflected shock wave if the mixing zone would interact with reflected shock wave. It will be accepted that the incident laser-light attenuation starting at the moments M_6, M_5 and M_3 demonstrate the existence of the mixing zone itself.

Ignition Experiments

The boundary between a small and a large activation energy was around 1 600 K in the temperature of reflected shock wave, or 1 ms in the ignition delay time. Base on the result of previous paragraph, the first rarefied wave reached to the position A after 1.22 ms from the arrival of incident shock wave. For a compression-ignition experiment with a longer ignition delay, the rarefied wave will propagate through the mixture near the end of the tube, in which preflame reactions are in progress. We have to examine the relation between the rarefied wave and position / time of the ignition to occur.

Pressure is recorded in positions A, B, C and D, and transmitted light is observed in positions C, E and F. Figure 4 shows a pressure trace of position A. Incident laser-light attenuation and pressure histories of position C are shown in Fig. 5. The temperature behind reflected shock wave is 1438 K, made it similar to the one of previous paragraph. As shown in Fig. 5, after the reflected shock wave passed at R_3, pressure has slightly fallen at the moment K_3, then at P_3 it started to rise, and finally rose at the time B_3. In the positions B and D, similar behaviors could be seen. The pressure rise such as started at P_3 could not be observed when the driven gas was pure argon; this pressure rise is not due to the effect of gas flow. This pressure rise could be seen using pure helium as a driver gas; it is not due to the ethanol which is contained in the driver gas.

Characteristic behaviors in waves and light-attenuation histories of an ignition experiment are shown as a time-to-distance diagram in Fig. 6. Also here we can find by the transmitted-light history the arrivals of contact surface and interference which is apparently the mixing region. Two rarefied waves are perceivable, which are caused by interactions with reflected shock wave and are propagating to the end of the tube.

The first pressure-rise timings P_2, P_3 and P_4 behind the reflected shock wave at the positions B, C and D can be tied together by a solid straight line. Another line would be drawn from the moment P_1 having a gradient of segment E_1-E_2, which will intersect a extrapolated segment P_3-P_2. We can get there a point Ip as an intersection. If the pressure rise come up from the time P_1 is based on the same phenomenon as the ones at P_2, P_3 and P_4, the origin of the phenomenon would be the time Ip. The wave E_2-E_1 is a rarefied wave. This rarefied wave has propagated through the mixture in which preflame reactions were in progress. It is considered that the preflame reactions were accelerated and then a pressure rise was caused under the effect of this rarefied wave.

A steep pressure rise of real ignition started at the time B_1 near the end of the tube and propagated toward the driver section. A trigger for the ignition is raised not at the end of the tube, but a little to the inside of the tube. It should be considered that the ignition delays shown in lower temperature side of the Arrhenius plots than the inflected point are not the pure chemical delays under the pure conditions of constant temperature and pressure, given from the speeds of incident shock wave.

Visualization, spectroscopic analysis of this phenomenon, and reasoning out of the ignition acceleration will appear in another paper of ours in the very near future.

Conclusion

The activation energy for the compression ignition of a methane mixture is reduced suddenly to the almost half when the temperature behind a reflected shock wave decreases down to 1 600 K and ignition delay becomes longer than 1 ms. We have examined, and reasoned this curious phenomenon. A mixing zone could be raised ahead of a contact surface probably at a rupture of a partition diaphragm. The mixing zone interacts with reflected shock wave, then a rarefied wave is initiated. When an ignition delay is longer than 1 ms, the rarefied wave propagates through the mixture in which preflame reactions are in progress. The preflame reactions would be accelerated and then a pressure rise will be caused under the effect of the rarefied wave. A steep pressure rise of real ignition starts near the end of the tube and propagates toward the driver section, but the ignition is originated not at the end of the tube, but a little to the inside of the tube.

The ignition delays in lower temperature than the critical point are not the pure chemical delays under the pure conditions of constant temperature and pressure, given from the speeds of incident shock wave.

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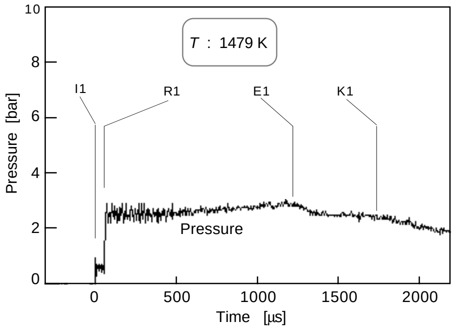

Fig. 1 Pressure trace of argon compression at position A: 17 mm distant from the end of the driven section, the nearest position to the end in this experimental set-up

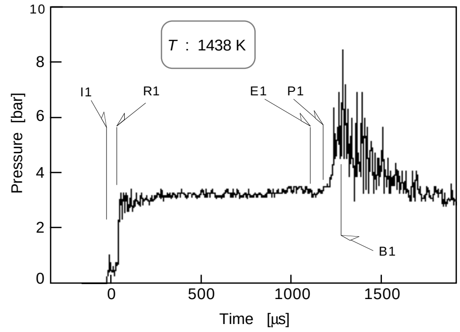

Fig. 4 Pressure trace of methane compression ignition at position A: the nearest point to tube end. Temperature behind reflected shock wave is 1438 K

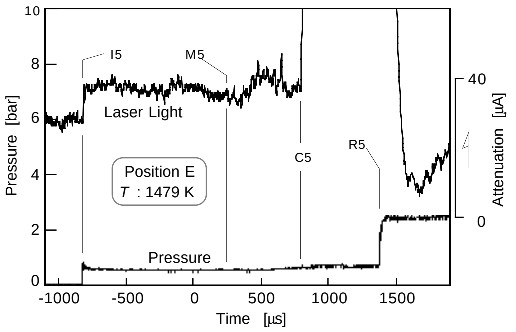

Fig. 2 Incident laser-light attenuation and pressure histories of argon compression at position E: 990 mm distant from the end of the driven section. In this figure the transmitted light trace swings upwards as the incident light is attenuated

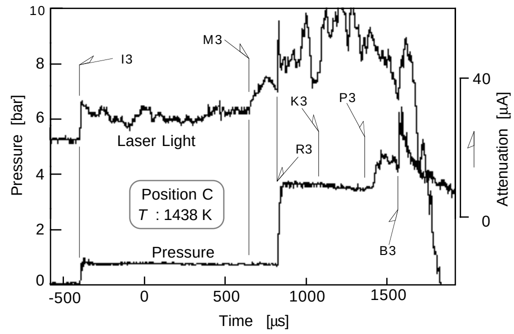

Fig. 5 Incident laser-light attenuation and pressure histories of methane compression ignition at position C: 340 mm distant from the end of the driven section

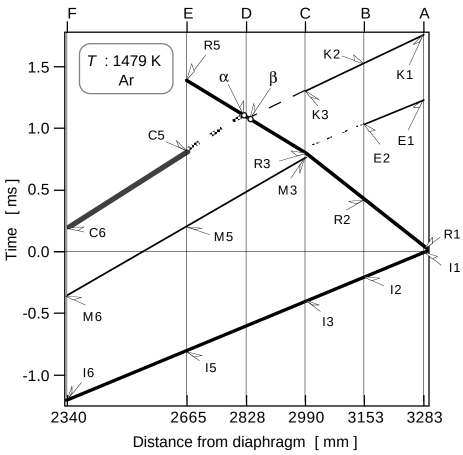

Fig. 3 Time-to-distance diagram of argon compression. Abscissa is the distance from the partition diaphragm membrane and the ordinate the elapsed time after the incident shock wave has arrived at the end of driven section

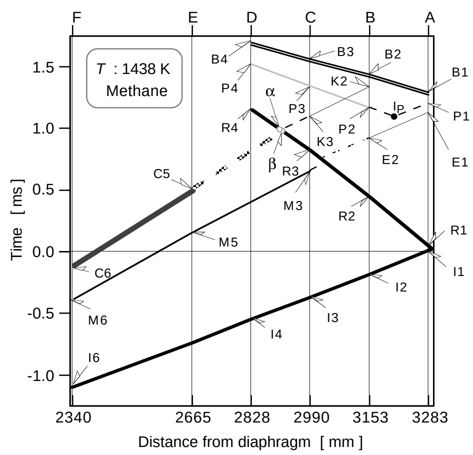

Fig. 6 Time-to-distance diagram of methane compression ignition to the temperature of 1438 K