A Study on Initiation Processes of Gaseous Detonation in Methane/Oxygen Mixtures*

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Initiation processes of gaseous detonation waves were observed experimentally in a tube. Gases to be tested were methane/oxygen mixtures of various initial compositions. The initiation was performed by blast waves caused by gaseous detonations of initiator gases which were stoichiometric oxyhydrogen mixtures at various initial pressures, the initiation energies of which were estimated by the method developed by the authors. The initiation energies were estimated from the experimental result of the decay of an inert planar blast wave produced by the initiator by utilizing the point source blast wave theory. The critical initiation energies were evaluated for the near limit composition of the mixture. In the initiation processes, local explosions were observed which played an important role under the critical condition, which was produced not only by the effect of the leading shock wave but also by that of the detonation products of the driver gas.

Key Words: Combustion, Detonation, Initiation Energy, Blast Waves, Methane

1. Introduction

The classical studies of detonation concerning detonability limits and initiation problems were almost always performed by using detonation tubes. The tubes usually consisted of a driver tube and a driven tube, the former being filled with a gas used as an initiating charge and the latter being filled with a gaseous mixture under test.

Gordon et al.⁽¹⁾ used a 20 mm diameter tube which was 12 m long. The length of the driver section was 2 m and the driver gas was stoichiometric oxyhydrogen mixture. The pressure and the amount of diluent of the driver gas as parameters were changed to observe their influences on the initiation processes. They found that the detonability limit would depend on the initial pressure and composition of the driver gas. They did not dispute on the initiation energy, but this observation clearly showed that the detonability limit was a

function of the initiation energy.

In recent years, the initiation of unconfined spherical detonations has attracted considerable attentions⁽²⁾⁻⁽⁴⁾. The importance of the critical initiation energy with regard to safety has been recognized by many researchers who have been engaged in determining critical initiation energies for various combustible mixtures which are used as our energy resources.

The initiation processes of detonation are not only determined by the initiation energy but also the rate of energy deposition per unit volume, which Lee et al.⁽⁵⁾ called a power density of the initiator. So, the critical initiation energies obtained by the various different methods, e.g., solid explosives, a laser spark, an exploding wire and a gaseous detonation, are quite different from each other. It is desirable to use the power density as an alternative to the initiation energy. However, it is very difficult to estimate exactly the power density for various types of initiator. An appropriate measure of the initiation energy is necessary. In refs. (6) and (7), the authors had proposed the method of evaluating the initiation energy as a universal parameter. In this method, the initiation energy was defined as an energy whose power density

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is infinite. It was determined by an experiment in which decay of an inert blast wave caused by the initiating charge was measured and compared with the theory based on the assumption that the energy was released at an infinite power density, i.e., the point source blast wave theory.

This method had been successfully applied to the evaluation of the critical initiation energy of the detonations in stoichiometric methane/oxygen/nitrogen mixtures as an amount of a diluent (nitrogen) as a parameter⁽⁶⁾.

The present paper describes the results of an experiment to observe initiation processes of planar detonation waves and to estimate the critical initiation energy of methane/oxygen mixtures of various initial compositions. Methane is one of the most important and fundamental of the gaseous fuels that we use. The initiating charge was same as that used in ref. (6) which was an oxyhydrogen mixture with the initial pressure being changed as a parameter.

The initiation processes were measured by pressure transducers and ionization probes embedded along the tube wall. The critical initiation energies were estimated for several near-limit compositions of the mixtures and compared to other experiments and calculations.

2. Experimental

In the present experiment, a linear detonation tube with a circular cross section was used. The diameter of the tube is 30 mm for both driver and driven sections. The tube is made of stainless steel. A schematic of the apparatus is shown in Fig. 1. The length of the driver section is 595 mm and that of the driven section (test section) is 3 495 mm and they are separated by Mylar sheet (0.03 mm thick). There are

Fig. 1 Schematic diagram of experimental apparatus.

11 measuring positions (called as MP.1 through 11 respectively) located 400, 700, 1 000, 1 300, 1 500, 1 800, 2 100, 2 400, 2 700, 3 000, and 3 300 mm downstream from MP. 0 which is located 100 mm upstream from the Mylar sheet. At each measuring position, pressure transducers (PCB 113A24) and ionization probes are be mounted on opposite sides of the tube wall to detect the shock and reaction fronts simultaneously. The signals from these sensors were stored by a 3-channel transient recorder with one microsecond resolvability. The signals from all ionization probes were collected and stored by one channel of the recorder and the pressure variations at two adjacent points were stored by the other two channels. At least eleven test runs must be performed for one condition. The reproducibility was confirmed by measuring twice at the same location and was good except at the location where a strong local explosion was generated. The propagation velocity of the shock wave was obtained from the known distance between the two adjacent pressure transducers and the corresponding elapsed time between the times when the shock front arrives at each measuring position.

The present experiment used stoichiometric oxyhydrogen mixtures with initial pressures p_d equal to 101.3, 79.97 and 53.33 kPa as driver gases. A Shchelkin spiral was inserted in the driver tube to ensure a transition to detonation of the initiating charge within a length equal to 595 mm. The details of the experimental method to evaluate the initiation energy are described in ref. (7). Table 1 shows the numerical values of the initiation energy E_0 , the characteristic distance R_0 , and the equivalent initiation energy for a spherical geometry E_0'' . The last quantity was defined as an energy which could produce a spherical blast wave whose characteristic distance equals that of the planar blast wave observed here. This method was proposed by Wolanski et al.⁽⁸⁾ and is important for this type of experiment because comparison should be made with the spherical or cylindrical experiments. About 30-40% of the chemical energy contained in the

Table 1 Initiation energy of driver used.

Driver gas	p_d (kPa)	E_0 (MJ/m ²)	R_0 (m)*	E_0'' (MJ)*
2H ₂ + O ₂	101.3	0.812	2.86	42.9
2H ₂ + O ₂	79.97	0.553	1.98	13.7
2H ₂ + O ₂	53.33	0.308	1.09	2.34

*) $R_0 = E_0 / (2\gamma p_0) = (E_0'' / (4\pi\gamma p_0))^{1/3}$, where γ is the ratio of specific heats and p_0 the initial pressure of the medium.

initiator was used to form a planar blast wave. The rest of the energy was lost in the form of various pressure waves like a detonation wave. The initiation energy is nearly proportional to the initial pressure.

In the present report, the tested mixtures were methane/oxygen mixtures of various compositions; in particular, in near-limit conditions with no diluent. The initial pressures of the tested gases were 99.99 kPa. The mixtures were prepared in mixing tanks and compositions were determined by adjusting the partial pressure of each component. Equivalence ratios of the mixtures were selected to cover the lean and rich detonability limit of the mixture.

3. Results and Discussion

A series of pressure traces at MP. 3 through MP. 7 is shown in Fig. 2 for the case that the initiator is an oxyhydrogen mixture with initial pressure equal to 53.33 kPa which produces a blast wave with initiation energy E_0 equal to 0.308 MJ/m^2 as was shown in Table 1, and that the tested mixture is a methane/oxygen mixture with equivalence ratio ϕ equal to 0.22 and initial pressure equal to 99.99 kPa. The dashed line denotes the trajectory of the reaction front. At MP. 3, the leading shock wave arrived at a time about 1.45 ms after the wave have passed MP. 0 and the reaction front at about 1.75 ms. About 2 ms, a gentle rise in the pressure appears and it grows to an explosion-like pressure wave at MP. 4. This local explosion wave has a front which moves forward to overtake the leading shock wave as it propagates downstream to MP. 5 and 6, while the reaction wave keeps a considerable distance behind the leading wave. Ignition may occur in the low density and high temperature region located some distance behind the leading shock wave. This ignition zone originates from the contact surface between the detonation products of

Fig. 2 Pressure variations at several measuring points. In this case the well-established detonation was not formed.

the driver gas and the gas mixture under test. The tested gas is successively ignited to produce small explosion waves which overtake the leading shock wave to prevent it from decaying. The new local explosion wave passes MP. 7 at about 0.5 ms after the leading wave has passed. No established detonation wave is formed in this case, but there is a possibility that these small explosions may occur as the wave propagates downstream to cause a well-established detonation wave. In any case, it is a near-critical condition.

In Fig. 3, a series of pressure waves for the condition under which the detonation wave was initiated in the tube is shown. The initiation energy is same as in the previous case but the equivalence ratio ϕ is increased to 0.35. Note that the scales of time and pressure are changed from those in Fig. 2. In this case, a strong ignition occurs at MP. 6 just behind the leading shock wave which immediately becomes an overdriven detonation wave. At MP. 7, the reaction front already overtakes the leading shock wave so that the wave is a well-established detonation wave and it decays to the Chapman-Jouguet detonation wave. The pressure oscillation is observed at MP. 4 and 5, and its amplitude decreases at MP. 6 where the strong ignition occurs. At MP. 7, the well-established detonation wave is formed and there is no pressure oscillation. It should be noted that the transverse waves appear only in the transitional stage. Moen et al.⁽⁹⁾ suggested that the effects of the wall confinement were crucial in this transitional stage. The local explosion wave is generated at a point, rather than at a plane in some cross section of the tube so that a spherical divergent shock wave may be generated. This causes the pressure oscillation. In Fig. 4, typical pressure traces are shown for the onset of spinlike detonation but this oscillation is also unstable to

Fig. 3 Pressure variations at several measuring points. In this case the well-established detonation was formed.

decay as the wave propagates downstream. It is interesting to note that a ratio of spin-pitch to tube diameter for this spin-like wave is 3.2 which is very close to the theoretical value 3.1 calculated by Fay⁽¹⁰⁾.

Figure 5 shows typical trajectories of the leading shock waves and the following reaction wave fronts for the cases that the equivalence ratio of the mixture is 0.22 and the initiation energies are 0.812 and 0.308 MJ/m². In the former case, the distance between the leading shock front and the reaction front is relatively small compared to that of the latter, and at the final measuring point they coincide with each other. In the latter case, the two wave fronts never coincide in this tube. However, there remains a possibility of coincidence if the tube were a little longer. The variation of the distance between the shock front and the reaction front is affected by the initiation energy because in the

Fig. 4 Pressure variations at several measuring points. In this case the spinlike wave was observed.

Fig. 5 Trajectories of leading shock waves and reaction fronts.

transitional stage, it is not only determined by the chemical process but also by the hydrodynamic structure behind the leading shock wave. For a large initiation energy, the detonation products of the driver gas have a large energy so that the contact surface between the high-temperature products and the unburned test gas penetrates deep inside the driven tube. The first ignition may occur near this contact surface and the reaction wave is thus propagated, generating the pressure waves shown in Fig. 2. This situation is analogous to that observed by Edwards et al.⁽¹¹⁾ in the experiment on the initiation of a detonation wave by an incident shock wave. Due to the effects of the pressure waves generated by the reaction wave, the leading shock wave is gradually strengthened and this causes the distance between the shock front and the reaction front to become smaller. At MP. 11 (3.4 m downstream from MP. 0), the reaction wave overtakes the leading wave and the detonation wave is established. For lower initiation energy, the local explosion wave is too weak to strengthen the leading shock wave and the overtaking of the leading wave by the reaction wave does not occur within the tube used. As mentioned above, the possibility of the formation of detonation wave still exists for this case if the tube were a little longer. But, considering the fact that the characteristic distance R_o of the initiator of $E_o = 0.308$ MJ/m² is equal to 1.09 m and that of $E_o = 0.812$ MJ/m² is equal to 2.86 m, the possibility of detonation may be very small for the case $E_o = 0.308$ MJ/m².

The variations of shock Mach numbers were shown in Fig. 6. The process of establishment of detonation consists of three stages. The first stage is a

Fig. 6 Variations of shock Mach number with propagation distance. Driver gas pressure is 101.3 kPa.

decaying stage of the initiating blast wave, the second is an acceleration stage to the overdriven detonation and the last stage is a decaying stage from the overdriven detonation to the Chapman-Jouguet detonation wave of which the propagation Mach number is calculated assuming the chemical equilibrium behind the detonation front and is shown by the horizontal dashed lines denoted CJ in the figure. For the fixed initiation energy, the appearance of the second stage becomes delayed as the equivalence ratio of the mixture departs from unity.

In observing the initiation processes, the critical initiation energies for several near limit mixtures were estimated. Figure 7 shows the obtained critical energy converted into spherical geometry as a function of the mixture equivalence ratio. In other words, the solid circles denote the averaged critical compositions of the several experiments being fixed the initiation energy. In the present method of initiation, the range of energy is not so wide, so it would not be possible to cover the whole range of the equivalence ratio of the mixture. The obtained critical energies were limited to the near-limit compositions. In this figure, the experimental results of Matsui et al.⁽¹²⁾ and Ubbelohde et al.⁽¹³⁾ as well as the calculated results of Westbrook et al.⁽¹⁴⁾ were also shown. In the lean limit, the present results agree well with the limit obtained by Ubbelohde et al. and it seems to be less affected by the initiation energy, while in the rich limit some differences do exist. In Ubbelohde's experiment, the driver gas was a stoichiometric oxyhydrogen mixture

at 101.3 kPa and the driver tube was 1 in (25.4 mm) diameter and 5 ft. (1.5 m) long so that the initiation energy of the initiator was two or three times larger than our strongest initiator. In addition, in the rich limit, the difference of 1% volume fraction causes that of 0.1 in the equivalence ratio, while in the lean limit the same difference in the volume fraction corresponds to that of 0.02.

The method of Matsui and Lee (i.e., the critical tube diameter method) as well as that of Westbrook (based on the Zeldovich criterion) to estimate the critical initiation energy are very powerful in covering almost the whole range of the equivalence ratio; however, in principle, they cannot be applied to the limiting compositions that are obtained by the present study.

4. Conclusions

The initiation processes of detonation waves in the mixtures of methane/oxygen at various compositions were observed and the following conclusions were derived:

(1) In near-critical conditions, small explosion waves are formed behind the leading shock wave to prevent it from decaying. The distance between the shock front and the reaction front keeps a considerable value in the tube.

(2) In supercritical conditions, the reaction waves overtake the shock front as a result of the occurrence of the strong explosion wave which initiate a transient, spinlike pressure oscillation behind the shock front.

(3) The critical initiation energies for several near-limit compositions are estimated by using the method of evaluating the initiation energy released by the present initiator, i.e., the oxyhydrogen detonation with the various initial pressure. The obtained limiting composition at lean condition agrees well with the result of Ubbelohde and it is less affected by the initiation energy, while in the rich limit some differences appear due to the effect of the initiation energy. The advantage of this direct method of obtaining the critical initiation energies in the near-limit mixtures is thus confirmed.

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