Mechanism of Explosion Induced by Contact of Hypergolic Liquids

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The nature and mechanisms of explosions caused by the contact of hypergolic liquid propellants were investigated in detail for several combinations of fuels and oxidizers. It is shown that the explosion phenomena observed can be classified into three categories. 1) In the case of N_2H_4/NTO, sudden gasification of a superheated liquid layer formed at the boundary of two liquids occurs spontaneously and a detonation-like reaction proceeds in the reactive mixture produced. 2) In the cases of MMH/NTO and UDMH/NTO, the sudden gasification is caused by the shock of a local ignition, and a turbulent-combustion reaction proceeds in the reactive mixture produced. 3) In the cases of hydrazine type fuels/FNA, the sudden gasification occurs spontaneously as in the case of N_2H_4/NTO, but it is not augmented by chemical reaction, and in these cases the observed explosion is weak. Information on the vapor layer, which is formed between reactive fuel droplet and pool liquid plays an important role for the occurrence of explosion, is also given, that is based on the high-speed motion picture records.

Introduction

THE phenomenon of explosion induced by the contact of hypergolic liquid propellants has potential importance for the combustion process in rocket engines and for safety procedures.[1] The authors suggested in Refs. 2 and 3 that the explosion in a N_2H_4/NTO system is caused by a sudden gasification of the superheated thin surface layer of the low boiling point NTO.

In the present investigation the nature and the mechanisms of explosion were investigated in detail for various combinations of hypergolic liquid propellants—fuels:hydrazine (N_2H_4), monomethylhydrazine (CH_3NHNH_2, MMH), unsymmetrical dimethylhydrazine ($(CH_3)_2NNH_2$, UDMH), and mixed fuel; and oxidizers:nitrogen tetroxide (N_2O_4, NTO) and fuming nitric acid (HNO_3, FNA). Information on the vapor layer, which is formed between reactive fuel droplet and pool liquid plays an important role for the occurrence of explosion, is given based on the high-speed motion picture records obtained using a specially devised method.

Experimental Procedure

A schematic diagram of the experimental apparatus is shown in Fig. 1. When the electromagnetic valve is opened, the fuel stored in the quartz pipe flows downward and a fuel droplet forms at the lower end of the pipe. The fuel droplet (droplet diameter, D) falls into an oxidizer pool (usually at the center of the pool, see Fig. 1a) placed in an anechoic chamber, with an impact velocity, V, based on the falling height. A pressure transducer (1.5 dB in the range of 20 Hz ~ 300 kHz), located 18 cm from the contact point of the droplet with the pool surface, receives the pressure waves emitted in the course of the phenomenon and two photodiodes receive the light emitted by chemical reaction.

It was confirmed by use of a thin thermocouple that the surface temperature of droplet or pool, before contact, is near to the wet bulb temperature of each liquid. In the course of these experiments the surrounding temperature was in the range of 18 ~ 20°C. The boiling point of the used fuels and oxidizers at atmospheric pressure are as follows: N_2H_4 = 114°C, MMH = 88°C, UDMH = 63°C, NTO = 21°C, FNA = 83°C.

The colliding system of droplet and pool of hypergolic liquid propellants is complicated optically. The usual shadowgraph technique (Fig. 1a) indicates only the depressed pool surface, when focused to the center of the axially symmetrical phenomenon, and the bottom of the submerged droplet cannot be seen on the photograph, disturbed by the curvatures of depressed pool surface. Figure 1b shows a specially devised piece of equipment for the observation of the vapor layer formed between the droplet and the pool surface. The edge of a thin stainless steel plate, which is installed on the edge of a glass cell, cuts a falling droplet in two. One part continues to fall along the inner surface of the glass cell. The aspect of the vapor layer formed between the submerged half droplet and depressed pool surface can be observed at the position of inner surface of the glass cell, which makes clear the contour of the droplet (mixed previously with fine silicon oxide powder) by use of a front light and the surface of depressed pool liquid by use of a back light.

Experimental Results and Discussion

Figure 2a and Fig. 2b show the variation of the probability of explosion with impact velocity, when various hydrazine-type fuel droplets fall into a pool of NTO or FNA. In the case of N_2H_4/NTO, no explosion is observed when the impact velocity is 1 m/s, after which the probability of explosion increase with impact velocity. In the cases of MMH, UDMH/

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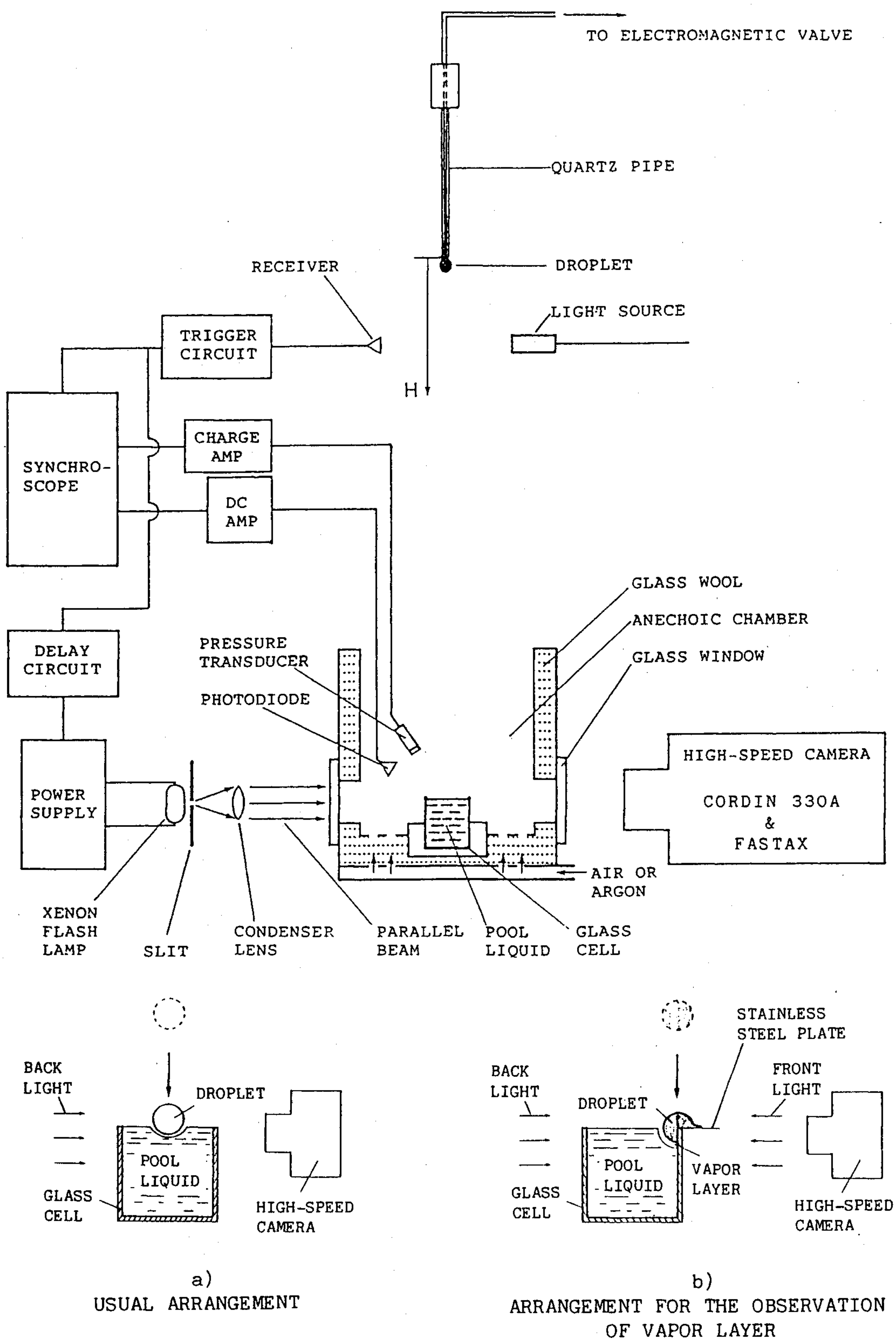

Fig. 1 Experimental apparatus.

NTO combinations, the probability of explosion is already 100% even at the low impact velocity of 1 m/s. It should be noted here, however, that the strength of explosion is highest in the case of N_2H_4/NTO. For the FNA oxidizer case, the probability of explosion is the largest for N_2H_4, less for MMH, and the least for UDMH.

Typical oscillograms of overpressures and emitted light, when various hydrazine-type fuel droplets fell into pools of NTO and FNA and caused explosion, are shown in Fig. 3 (D = 4.0 mm; V = 2.0 m/s). In the figure, the overpressure trace is shifted to the left, corresponding to the travel time of pressure wave between the impact point and the position of the pressure transducer, in order to compare the time of occurrence of explosion with that of light emission. The oscillogram (Fig. 3a) for N_2H_4/NTO combination shows that the explosion in this case is accompanied by simultaneous light emission, and the peak overpressure of explosion (P_{EX}) is very high. In the cases of MMH/NTO or UDMH/NTO com-

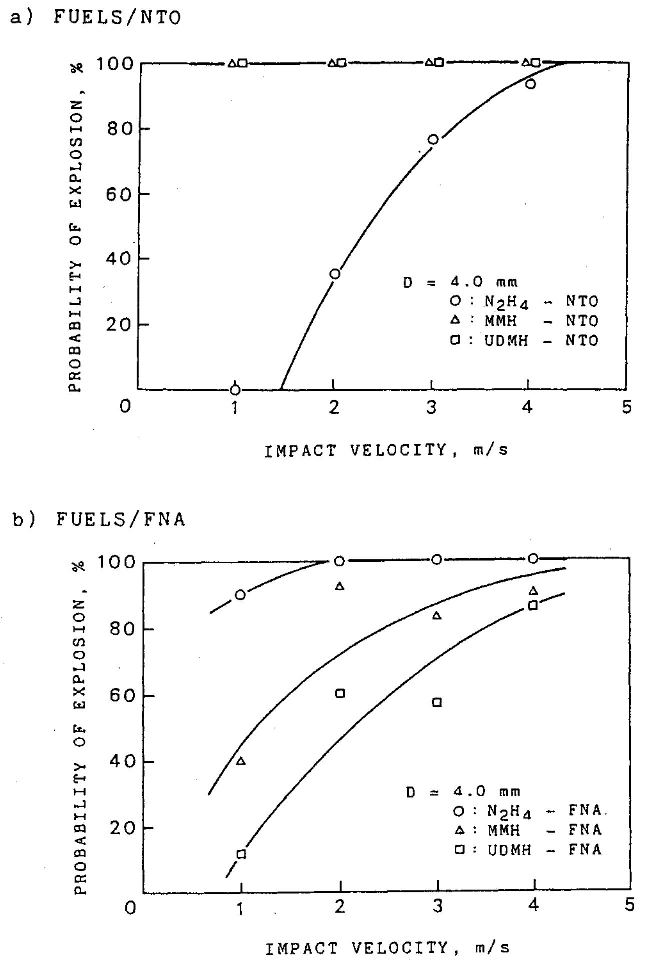

Fig. 2 Influence of impact velocity on the probability of explosion, D = 4.0 mm.

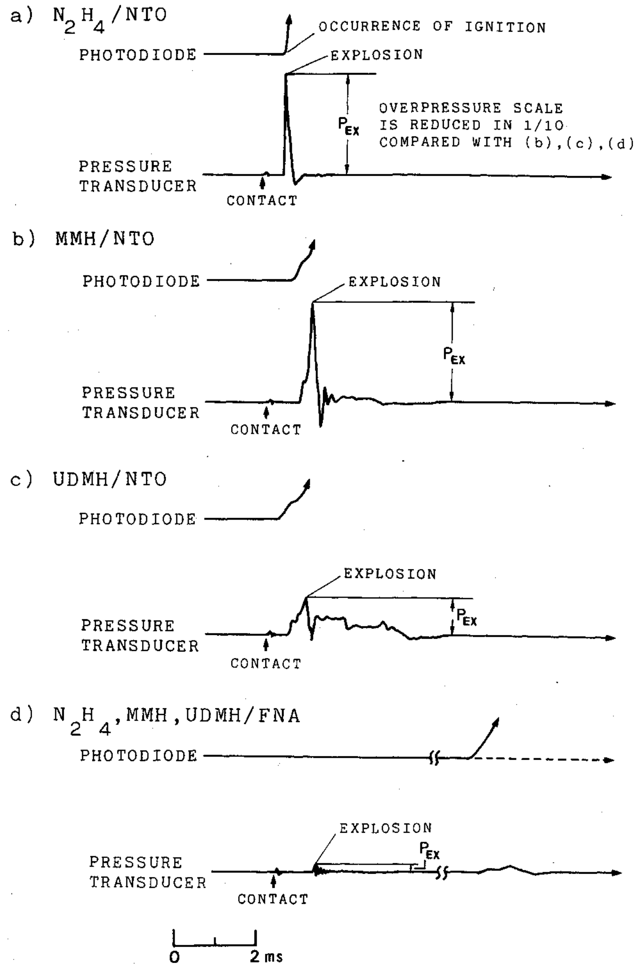

Fig. 3 Typical oscillograms of overpressure and emitted light, D = 4.0 mm, V = 2.0 m/s.

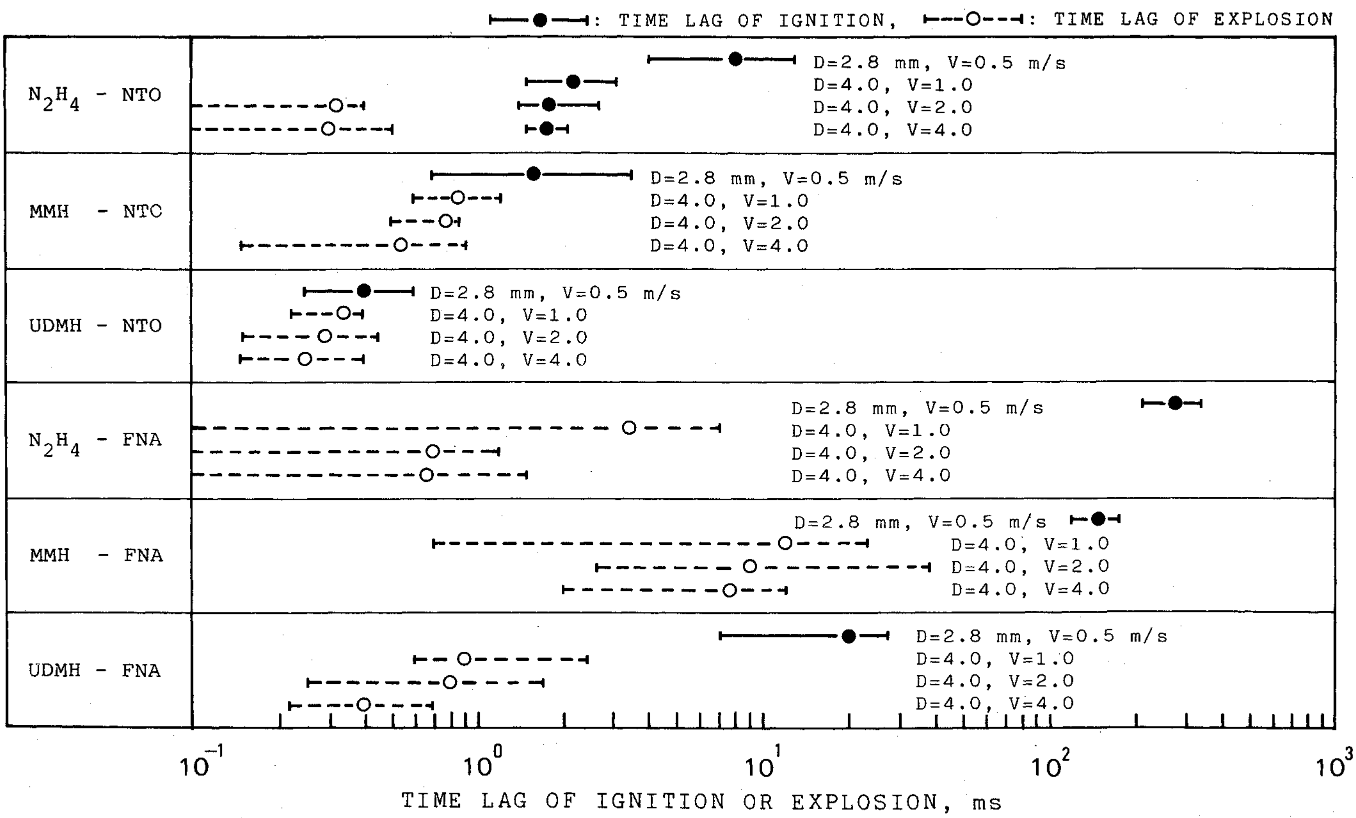

Fig. 4 Typical examples of distributions of the ignition or explosion time lags.

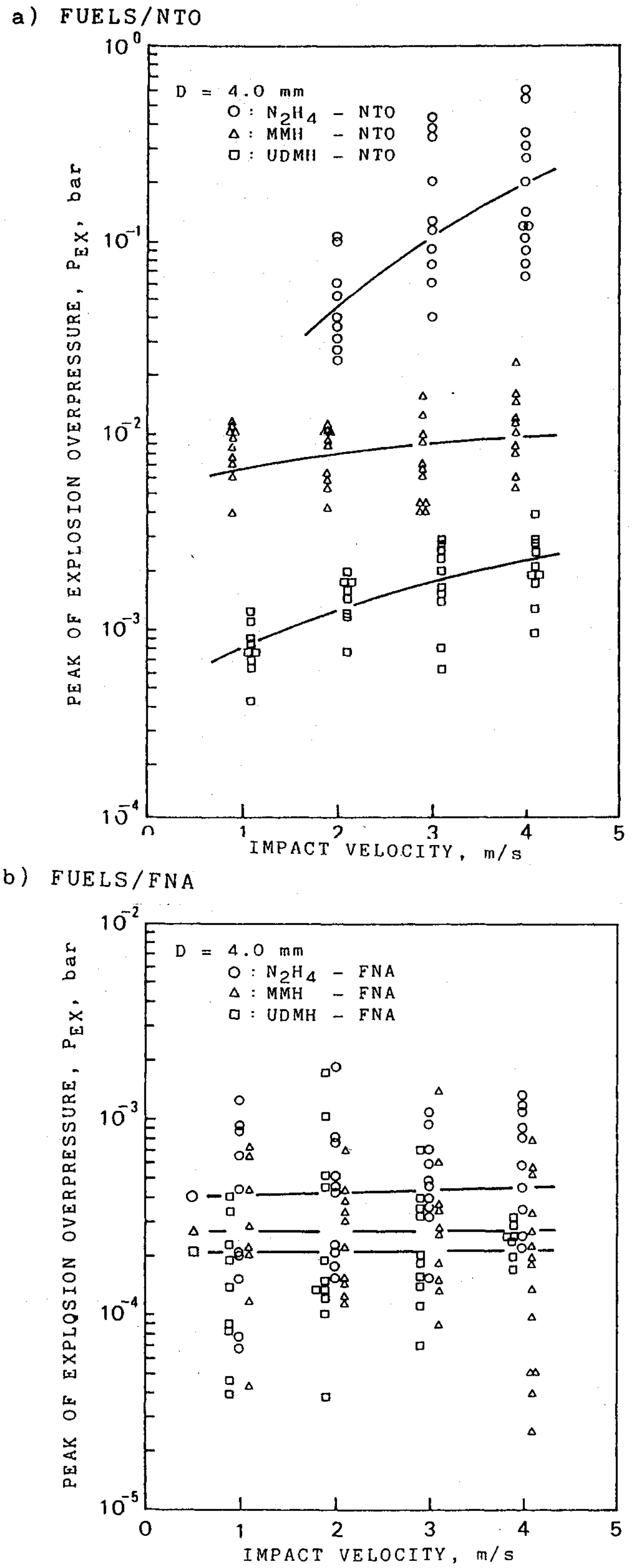

Fig. 5 Influence of impact velocity on the peak value of explosion overpressure (P_{EX}), D = 4.0 mm.

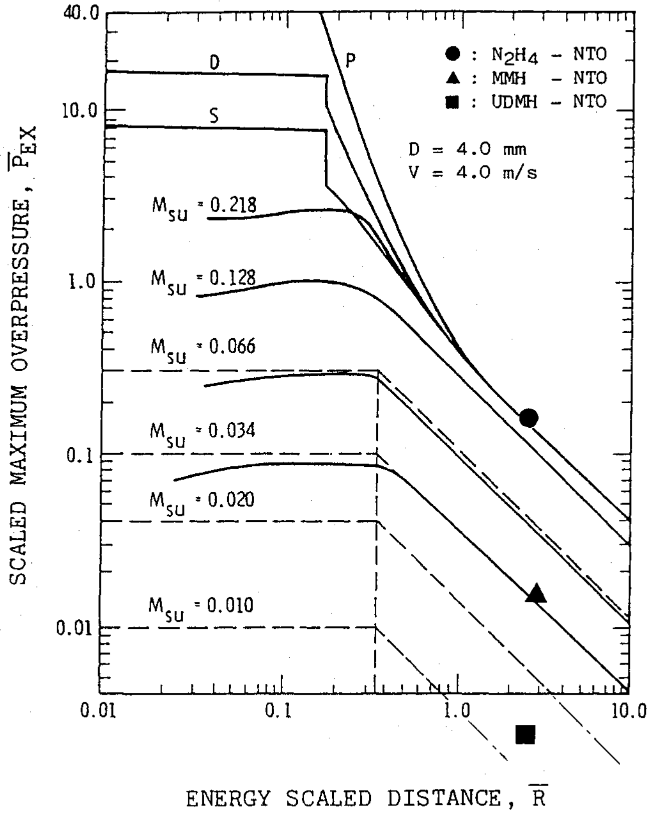

Fig. 6 Nondimensional maximum wave overpressure vs energy scaled distance for deflagrative explosion. M_{SU}: normal burning velocity/velocity of sound, P: pentolite, D: detonation, S: sphere burst.

binations (Fig. 3b or Fig. 3c), the light emission is observed before anything else and after 0.1 ~ 0.2 ms the explosion overpressure appears. The peak overpressure is one order or more lower than that in the case of N_2H_4/NTO combination. It is also seen in these oscillograms that the durations of pressure waves are in the order: N_2H_4 < MMH < UDMH. In the cases of fuels/FNA combinations, no light emission is observed on the occurrence of explosion and the peak overpressure is low.

For the mixed fuel of N_2H_4 and UDMH, an extensive experiment was conducted, changing the ratio of constituents in the fuel. The pool liquid is NTO as in the case of experiments shown in Fig. 3, droplet diameter is kept 4.0 mm and impact velocity, 4.0 m/s. When the weight percent of UDMH in the mixed fuel is increased from zero, it was found that until about 20% of all oscillograms observed kept the characteristics in the case of N_2H_4/NTO combination—very high explosion peak pressure and simultaneous light emission (Fig. 3a). In the range more than about 40%, all oscillograms obtained kept the characteristics in the case of UDMH/NTO combination—moderate explosion peak pressure and light emission advanced the appearance of the peak pressure (Fig. 3b and Fig. 3c). In the range of 20 ~ 40%, the oscillograms take the form either of those in the cases of N_2H_4/NTO or UDMH/NTO, and any intermediate forms of these two cases do not appear.

On the overpressure traces, as shown in Fig. 3, the explosion time lag (the time between the contact of the two liquids and the occurrence of explosion) can be evaluated. Also in the case where no explosion is observed, the ignition time lag (the time between the contact and the occurrence of usual ignition) can be evaluated by referring to the overpressure and light emission traces. Figure 4 shows the influence of droplet diameter and impact velocity on the ignition or explosion time lags. The time lags of ignition for the fuels/NTO combinations are shorter than those for the fuels/FNA combinations. The time lags of ignition for the same oxidizer are in the order: UDMH < MMH < N_2H_4. Furthermore, it is seen that the time lag of ignition decreases with increasing droplet diameter and impact velocity as described in Ref. 1. Also seen in the figure is that in the case of N_2H_4/NTO combination there is considerable difference in the region of appearance of the ignition and that of the explosion, at the droplet diameter of 4.0 mm and impact velocity of 4.0 m/s. However, in the cases of MMH, UDMH/NTO combinations, the regions of appearance of the explosions already overlap

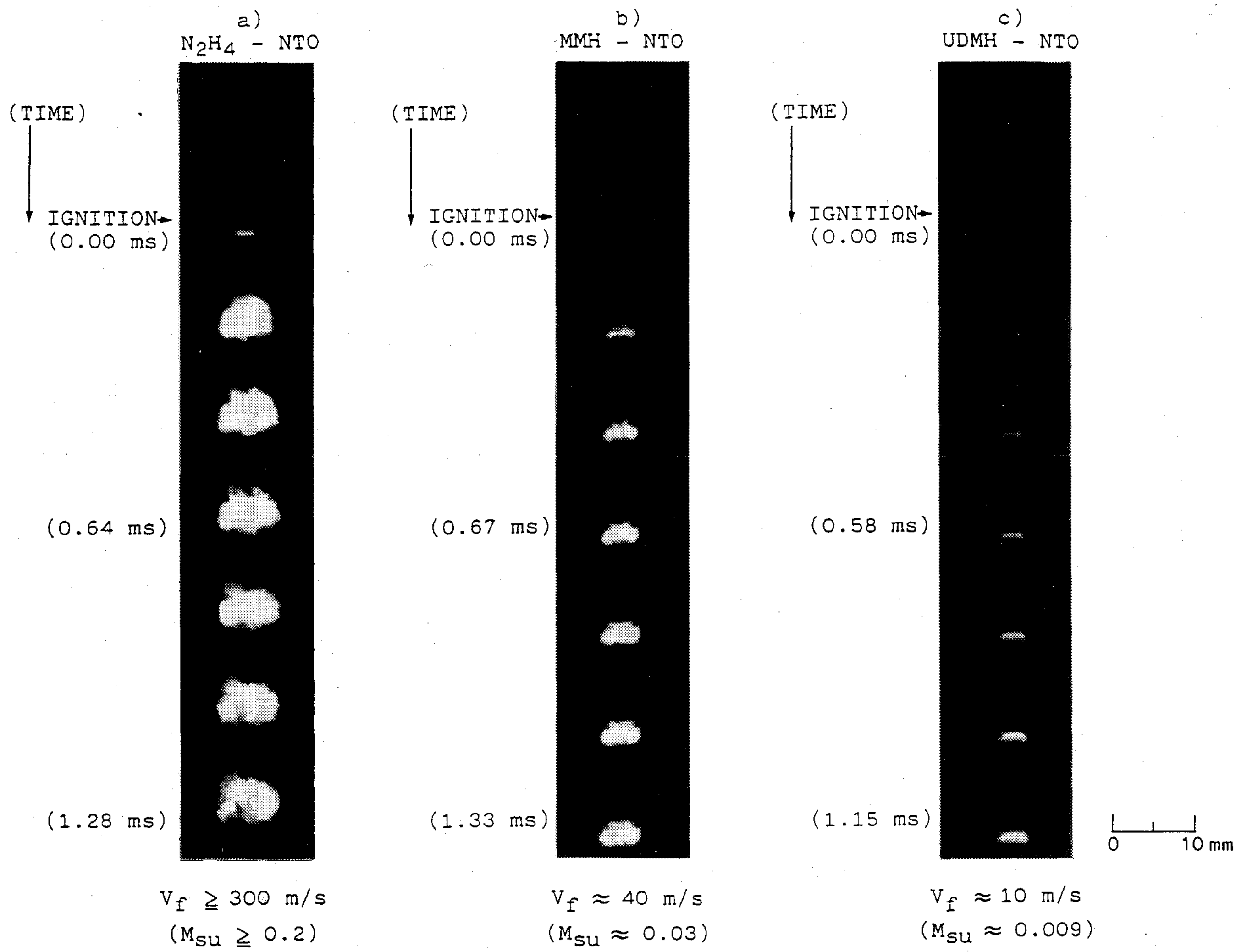

Fig. 7 High-speed motion picture records of expanding fire ball in the cases where explosion were observed: a) N_2H_4-NTO (4700 frames/s), b) MMH-NTO (4500 frames/s), c) UDMH-NTO (5200 frames/s). V_f: frame propagating velocity, M_{SU}: normal burning velocity/velocity of sound.

with those of the ignitions at the small droplet diameter of 2.8 mm and the low impact velocity of 0.5 m/s. This fact suggests the possibility of the explosion triggered by ignition, as shown in Figs. 3a and 3c.

Figures 5a or 5b shows the change of the peak of explosion overpressure, which was observed at a distance of 18 cm from the contact point, with impact velocity, for the combinations of various hydrazine-type fuels with NTO or FNA. In the case of NTO oxidizer, the strength of explosion increases with impact velocity. There is considerable difference in the strength between fuels. In the case of FNA oxidizer, however, the strength is kept nearly constant for the change of impact velocity. There is little difference in the strength between fuels.

In the previous experiment for a hydrazine droplet and nitrogen tetroxide pool, the volume of NTO which gasifies suddenly just before the explosion,[2] was evaluated based on the high-speed motion picture records. It was shown that for the case of $D = 4.0$ mm and $V = 4.0$ m/s, the volume is about 2.5 mm^3. In the present study, it was confirmed that for the same conditions of D and V nearly the same volume of NTO gasifies suddenly in the cases of MMH/NTO or UDMH/NTO explosions and that the magnitude of the source energy E is about 32 J.

An experimental and theoretical study of the effect of normal burning velocity on the blast wave produced by central ignition of a spherical cloud has been performed and the results are arranged in the graph of Fig. 6.[4] In the graph the maximum wave overpressure ($\bar{P}_{EX} = (P_{EX} - P_0)/P_0$) is plotted against energy scaled distance ($\bar{R} = R(P_0/E)^{1/3}$). The curve labeled P is experimental results for pentolite. All other solid curves are calculated ones. The curve labeled D is for detonation and S is for a sphere burst of the same energy density. The dashed lines represent the calculated overpressures for low-velocity flames. In the Fig. 6 graph, the observed peaks of explosion overpressure (averaged ≈15 data) for the cases of N_2H_4, MMM, UDMH/NTO combinations are plotted. The results of the plot suggest that in the case of the explosion of N_2H_4/NTO combination, a detonation-like reaction proceeds. In the cases of the explosions of MMH, UDMH/NTO combinations, a turbulent combustion-like reaction proceeds in the reactive mixture formed by the sudden gasification of NTO. This conclusion is compatible with the high-speed motion picture records of the growth of fire ball shown in Fig. 7, which indicate that in the case of N_2H_4/NTO, the flame propagating velocity is more than 300 m/s. In the cases of MMH/NTO and UDMH/NTO, the flame propagating velocities are ≈40 m/s and ≈10 m/s, respectively.

For a blast wave caused by an explosion, specific impulse i is also a significant parameter, and it is defined as

$$i = \int_{t_a}^{t_b} [p(t) - P_0]\, dt$$

where $[p(t) - p_0]$ is explosion overpressure, t_a is the arrival time of the blast wave to a position, and $t_b - t_a$ is the duration of the wave observed at the position. Figures 8a and 8b show the change of the specific impulse of blast waves observed at the distance of 18 cm from the contact point, with impact velocity for the combinations of various hydrazine-type fuels with NTO and FNA, respectively. It is shown that the mag-

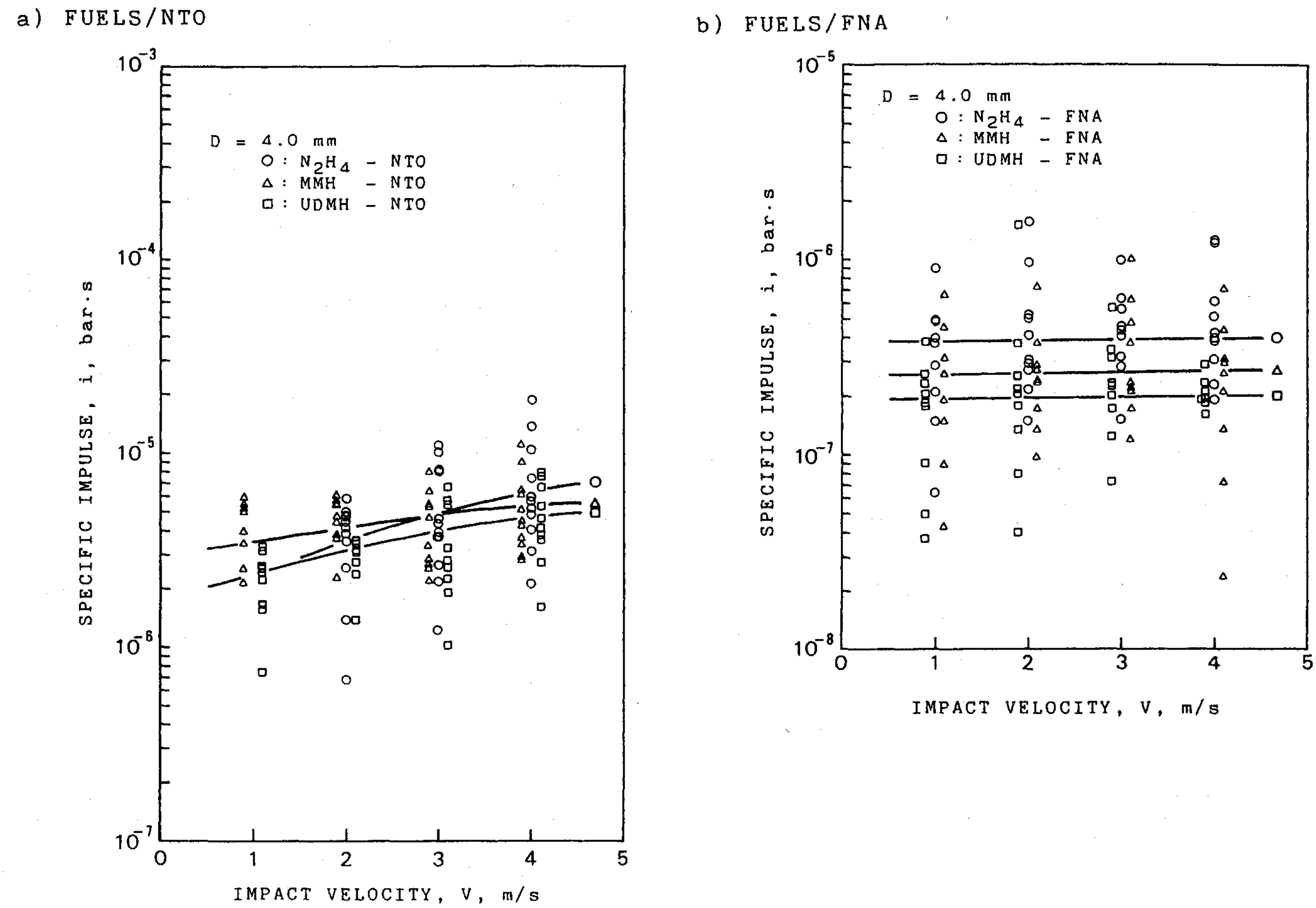

Fig. 8 Influence of impact velocity on the specific impulse of blast wave (i), D = 4.0 mm.

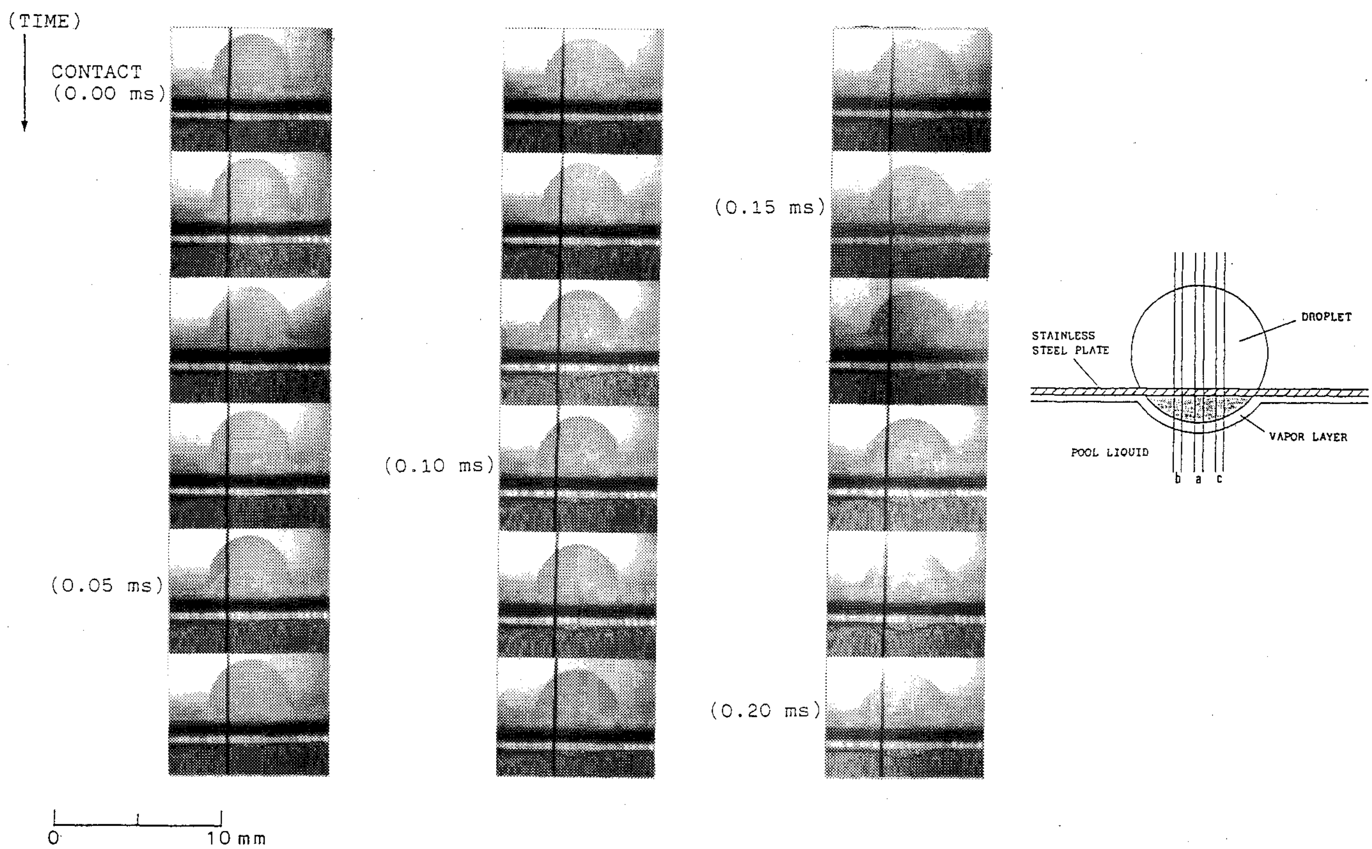

Fig. 9 High-speed motion picture records showing the change of the thickness of vapor layer, N_2H_4 droplet and NTO pool, D = 5.0 mm, V = 3.5 m/s, 89,000 frames/s.

nitudes of specific impulse for N_2H_4/NTO explosion, MMH/NTO explosion, and UDMH/NTO explosion are nearly the same regardless of the difference of heat release rate at the sources. For the value of specific impulse 5×10^{-6} bar · s, observed at the distance of 18 cm from the source in the case of hemispherical blast wave, the source energy is given as 30 J based on a blast wave theory in the far field,[4] and it agrees reasonably with the value of source energy evaluated formerly from the high-speed motion picture records (32 J).

The vapor layer formed between droplet and pool surface was observed using the specially devised method described previously. Figure 9 shows the high-speed motion picture records indicating the change of the vapor layer with the lapse of time for the case of N_2H_4 droplet and NTO pool. It was generally observed that the thickness of the vapor layer increases rapidly after the contact of the two liquids. The thickness of the vapor layer was determined by averaging the values measured at the three points indicated in Fig. 9. It was shown that its initial value (0.12 mm ~ 0.58 mm) is in the order: MMH/NTO ≈ UDMH/NTO > N_2H_4/NTO > UDMH/FNA > MMH/FNA > N_2H_4/FNA. Its growth with time (2.9×10^1 mm/s ~ 1.8×10^2 mm/s) is in the order: MMH/NTO > UDMH/NTO ≈ MMH/FNS ≈ UDMH/FNA > N_2H_4/NTO ≈ N_2H_4/FNA, under the condition of $D = 2.8$ mm, $V = 0.5$ m/s. It was also shown that the increase of droplet diameter or impact velocity has the tendency to decrease the initial thickness and to increase its increasing rate with time.

As for the relation of occurrence of explosion and growth of the vapor layer, it was confirmed generally that the occurrence is limited in the initial period where the thickness of the vapor layer is relatively small, i.e., less than ≈0.22 mm in the case of N_2H_4/NTO. Further detailed examination of the growing vapor layer, based on high-speed motion picture records, showed that the thickness oscillates with the frequency of 5 ~ 30 kHz as it increases with the lapse of time (the frequency depends on droplet diameter, impact velocity and the combination of fuel and oxidizer). The explosion occurs at the thickening phase of the oscillation in which the pressure in the vapor layer is lowered and the possibility for sudden gasification of the superheated liquid-layer in contact with it is large.

Conclusions

The nature and the mechanisms of explosion were investigated in detail for various combinations of hypergolic propellants—fuels: N_2H_4, MMH, UDMH, and mixed fuel; oxidizers: NTO and FNA. Comparing the experimental results obtained from pressure transducers and photodiodes and high-speed motion picture records with blast wave theories, it was concluded that the explosion phenomena are classified into three distinct categories: 1) the trigger for explosion, which is sudden gasification of the superheated surface layer of the lower boiling point liquid, occurs spontaneously, a detonation-like reaction proceeds in the reactive mixture of the source and the peak of explosion overpressure observed is very high (N_2H_4/NTO); 2) the trigger for explosion is caused by the shock of local ignition and a turbulent-combustion reaction proceeds in the reactive mixture of the source (MMH, UDMH/NTO); and 3) sudden gasification occurs spontaneously as in the case of N_2H_4/NTO, but it is not augmented by chemical reaction (fuels/FNA)—weak explosion.

It was also confirmed that the explosion observed in the case of the contact of mixed fuel (N_2H_4 + UDMH) and NTO belongs to the Type 2, described above, when the weight percent of UDMH in the mixed fuel is more than about 40%. This experimental result coincides with the practical experience in rocket technology that A-50 (50% N_2H_4 + 50% UDMH) is a reliable fuel combined with NTO in the point of the smoothness of combustion.

Information on the vapor layer, which is formed between reactive fuel droplet and pool liquid and plays an important role for the occurrence of explosion was also given, is based on the high-speed motion picture records. It was shown that the occurrence of explosion is limited in the initial period where the thickness of the vapor layer is relatively small and, in more detail, at the thickening phase of the vapor layer oscillation.

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References

[1]Friedman, R., Barnes, W. P., and Markels, Jr., M., "A Study of Explosions Induced by Contact of Hydrazine-type Fuels with Nitrogen Tetroxide," Technical Documentary Report ASD-TDR-62-685, 1962.

[2]Daimon, W., Tanaka, M., and Kimura, I., "The Mechanisms of Explosions Induced by Contact of Hypergolic Liquid Propellants, Hydrazine and Nitrogen Tetroxide," Twentieth Symposium (International) on Combustion, The Combustion Institute, Ann Arbor, MI, 1984, pp. 2065–2071.

[3]Tanaka, M., Daimon, W., and Kimura, I., "Explosion Phenomenon from Contact of Hypergolic Liquids," *Journal of Propulsion and Power*, Vol. 1, No. 4, 1985, pp. 314–316.

[4]Baker, W. E., Cox, P. A., Westine, P. S., Kulesz, J. J., and Strehlow, R. A., "Fundamental Studies in Engineering 5–Explosion Hazards and Evaluation," Elsevier Scientific Publishing Company, New York, 1983, Chap. 2.