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Experimental Study on the Combustion and Microexplosion of Freely Falling Gelled Unsymmetrical Dimethylhydrazine (UDMH) Fuel Droplets

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Abstract: The increasing demand for high energy density fuels and the concern for their safety have propelled research in the field of gelled propellants, where understanding the combustion of single gelled fuel droplets is the first stage to predict the spray combustion characteristics. The experiments utilized single-isolated freely falling gelled unsymmetrical dimethylhydrazine (UDMH) droplets instead of the conventional suspended droplet approach, in order to eliminate the perturbation associated with the suspension mechanism. Morphological transformations of the gelled droplet involved in the combustion processes were monitored by employing a high-speed digital camera, while the effects of ambient pressure and oxygen concentration on burning rate constants were also studied. The experimental results show that four main phenomena (droplet deformation, bubble formation and growth, vapor jetting and luminous jetting flame with “horn” shape) and three distinct phases were identified in the droplet combustion process; the high yield stress and polymer chain structure of polymer gellant are responsible for the appearance of bubbles with almost the same order of magnitude as the droplets. Increasing the ambient pressure can increase the burning rate, postpone the appearance of microexplosions, and weaken microexplosion intensity; while increasing the ambient oxygen concentration can promote the appearance of microexplosions, strengthen microexplosion intensity and increase the burning rate.

Keywords: gelled UDMH fuel; droplet combustion; microexplosion; burning rate constant

1. Introduction

Gelled propellants offer good storability without sacrificing high energy performance and operability for rocket applications, and are being considered for certain future rocket and atmospheric propulsion applications such as tactical missiles, ramjets, *etc.* [1]. The study of the combustion mechanisms of single droplets is necessary for characterizing and understanding the phenomena of spray vaporization and combustion. Liquid propellant droplets have been investigated extensively and comprehensive data have been published [2]. However, the ignition and combustion characteristics might be different, because of the influence of the gelling agent on the physical and chemical behavior of gelled fuel, so it is essential to conduct experimental and theoretical studies on the combustion of gelled fuel droplets.

Gels are multi-component propellants, and it will burn like a multi-component fuel. Extensive theoretical and experimental work has been carried out in the field of multi-component droplet evaporation/burning to understand the combustion mechanisms. Solomon and Natan presented the multicomponent droplet combustion [3,4]. Then, Ghassemi *et al.* studied the single droplet evaporation at elevated pressure and temperature [5]. The results showed a three-staged evaporation: (1) a period of more volatile component evaporation; (2) the period of increasing droplet temperature and almost no evaporation; (3) a quasi linear evaporation period. They also observed microexplosions at low pressure, but no microexplosions at high pressure.

The first study of combustion of an inorganic gellant fuel was conducted by Nachmoni and Natan [6,7]. Then, Arnold and Anderson studied the combustion process of inorganic silica gellant, JP-8-based gelled fuels droplets [8,9]. The experiments showed that the gelling agent had a considerable influence on the typical d^2 -law; a higher amount of gellant agent led to a reduced burning rate, and the droplet size as well as the droplet temperature during combustion depended strongly on the consistency of the gels.

Compared to the inorganic silica gellant, an organic gellant can burn with the base fuel. Nachmoni *et al.* were the first to conduct an experimental study on the combustion characteristics of organic gellant, non-metallized, JP-5-based gelled fuels (the droplet was suspended on the Pyrex wire) [6]. The experimental results indicated that gelled fuels had lower burning rate than pure liquid fuel of the same type, that radius changes obey the d^2 -law, and burning rate increased with increasing oxygen mass fraction and pressure. Recently, Solomon and Natan have carried out experimental work on the combustion mechanisms of organic gelled propellant droplets [4,10], where the phenomena during the combustion process were similar to those seen with multi-component droplets, and showed two stages: (1) a period of gellant formation; (2) a period of fuel vapor bubble formation and rupture. The process repeated itself until complete consumption of the base fuel and gellant. The results confirmed that the phenomenon of phase separation is not unique to gels and can also appear in multi-component fuels. They also compared the combustion behavior between the sub-critical and the super-critical pressure regimes, and the results showed that these combustion behaviors were different. Based on the experimental evidence of the combustion characteristics of organic gelled droplet, Kunin *et al.* developed a time-dependent, two-stage combustion model for organic gellant gelled droplets [11–13]. According to this model, the evaporation rate of the liquid fuel from the gelled droplet surface depends on the droplet size and significantly affects the thickness of the gellant layer. The stage during which

the gellant layer is formed is almost three orders of magnitude longer than the stage of bubble formation and layer rupture. Another study conducted by Mishra *et al.* [14,15] investigated the effects of gellant concentration, initial droplet diameter, and environmental pressure on the burning characteristics and flame structure.

Another type of gelled propellant are the so-called slurry fuels, which are mixtures of liquid fuel and solid particles, that were under serious consideration as high-energy fuels a few decades ago and can be advantageous, mainly in atmospheric propulsion systems [11]. Antaki presented the first theoretical model for the transient processes in the rigid slurry droplet during liquid vaporization and combustion [16]. He derived that the motion of the regressing surface constitutes a “ d^3 -law” (the diameter of the inner sphere decreases cubically with time). Recent advances in nanoscience and nanotechnology has enabled production, control, and characterization of nanoscale energetic materials, which have shown tremendous advantages over micron-sized materials. Yanan and Qiao conducted a series of experimental studies on the combustion characteristics of liquid fuel droplets with additional nano- and micro-sized aluminum particles [17–19]; the results indicated five distinctive stages (preheating and ignition, classical combustion, microexplosion, surfactant flame and aluminum droplet flame) for the *n*-decane/nano-Al droplet, they also carried out theoretical analysis to understand the effects of particle size on particle collision mechanisms and aggregation rate.

The objectives of the present study were: (1) to investigate the morphological changes of polymer gelled UDMH droplets during the overall combustion process; and (2) to explore the effects of ambient pressure and oxygen fraction on burning rate and microexplosions. The droplet combustion experiment and diagnostic methods are described. Several distinctive combustion stages are identified for freely falling droplets and droplets on a hot plate. Furthermore, theoretical results of burning rate are provided and compared with experimental results, and the differences between theoretical and experimental results are analyzed.

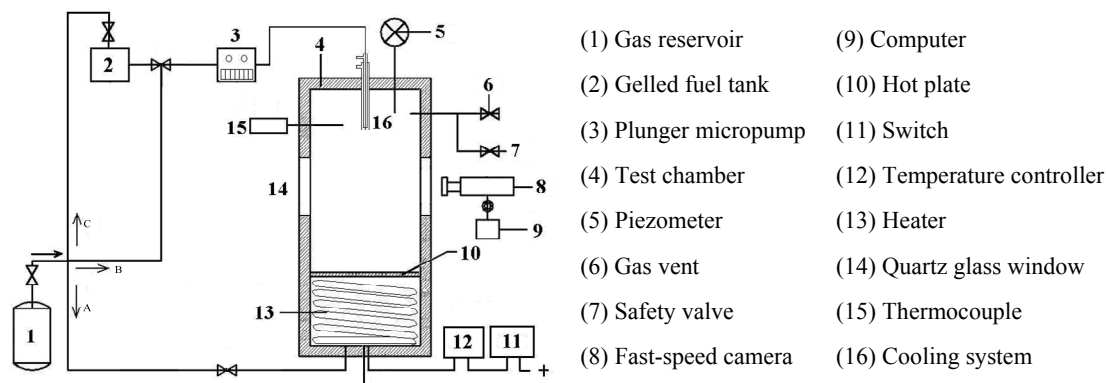
2. Experimental Setup

A schematic diagram of the experimental setup is shown in Figure 1. It consists of a high pressure chamber with a maximum pressure capacity of 2.5 MPa. The heating subsystem utilizes a temperature controller to control the chamber temperature; a 220 V AC power supply is used as the power for the heater coil. A plunger micropump which supplies a minimum volume 0.623 μL every time is used for generating single droplets. The diameter range of droplets is about 1–3 mm, and the maximum pressure of the pump outlet is 3.8 MPa. Burning processes are recorded by a high-speed (up to 120,000 frame/s) digital camera (Photron FASTCAM-ultima APX camera) with a speed of 4000 fps and a resolution of 512×1024 pixels. The height of the optical windows is 100 mm, and the images are analyzed by Image Lab software.

UDMH is used as the base fluid, and hydroxypropyl cellulose (HPC) is used as the gellant, whose maximum concentration is 3.0 wt%. The test chamber is purged firstly by injecting nitrogen gas, which replaces the air inside the chamber, then oxygen gas is injected. By changing the pressure of the test chamber before injection of oxygen gas, and after the injection, different experimental ambient conditions can be obtained. The heating subsystem can heat the gas in the test chamber up to 600 K (the thermocouple is located at the outlet of the droplet generator), and the droplets can be ignited

automatically for every experimental condition. Around 5–6 tests are conducted to improve the accuracy.

Figure 1. Schematic of the experimental apparatus.



For liquid fuels, plunger micropumps are widely used for generating single droplet, however, due to the high viscosity of gelled fuel, the droplet generating system may be blocked. Since gelled fuel is a shear-thinning non-Newtonian fluid, a spinning impeller is placed inside the tank to ensure that the gelled fuel fed will be from the plunger micropump to the test chamber. If the supplied volume of gelled fuel is constant every time, and the gelled fuel is stirred vigorously, the diameter of droplets can be seen as a constant.

The uncertainty within the ambient pressure measurement is ± 0.02 MPa. Due to the characteristics of the gelled fuel and normal gravity conditions, the droplets are not always spherical. A mean droplet diameter is determined from surface area and then converted to an equivalent droplet diameter. Uncertainty in the determination of droplet diameter comes from two sources. One is the scale factor, which is used to convert the droplet diameter size in terms of pixels to real units. A metal needle with a known diameter (2 mm) is used as reference scale. At the worst case, the measurements over a wide range of light intensities show that error is less than ± 0.02 mm. Another source for error in measuring droplet diameter is the optical effect of the hot and dense environment. Considering the density gradient around droplet and droplet falling, the range of errors for droplets is well within $\pm 5\%$ for the measuring technique used. Each of the measurements for the burning rates is obtained after an average of around 5–6 tests; the errors obtained in the values of the burning rate constant are in the range of $\pm 4\%$.

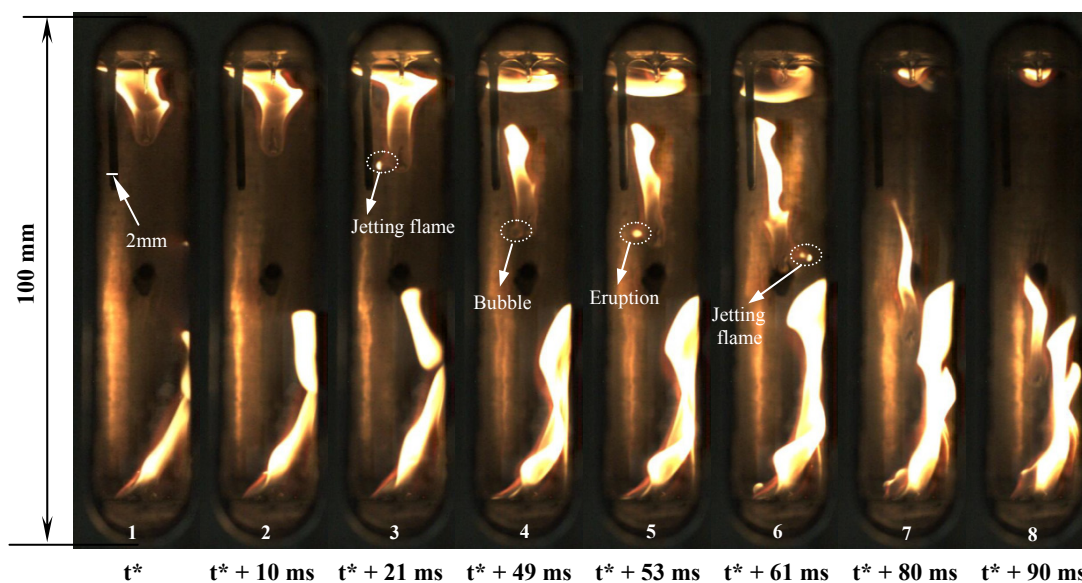
3. Results and Discussion

3.1. Freely Falling Gelled UDMH Droplets

The morphological evolution of flames and droplets can be observed clearly from progressive photographs during time evolution of burning freely falling droplets as shown in Figure 2. The conditions of pressure, temperature and oxygen mole fraction of the experimental chamber are 0.5 MPa, 473 K and 50%, respectively. Gelled droplets enter the experimental chamber, undergo sensible heating up to the autoignition temperature and then are ignited, and fall freely under normal gravity (the bottom flame is a result of the droplet burning after hitting the bottom surface of

experimental chamber, the top flame is a result of the remaining gelled fuel burning after droplet falling). As the UDMH evaporates from the outer part of the droplet, the gellant concentration increases on the surface (Frames 1–2). At a time $t = t^* + 21$ ms, a spontaneous UDMH fuel vapor eruption can be seen at the bottom of the freely falling droplet, leading to a luminous jetting flame with a “horn” shape (Frame 3), primarily due to the intensified heat transfer. About 49 ms later, a large bubble with a very thin outer layer, which is almost the same order of magnitude as the droplet, appears at the bottom of the droplet (Frame 4); and the bubble formation is different from other gels that were investigated by Natan [10] and Mishra [15]. Approximately 4 ms later the bubble ruptures; ignition of the ejected UDMH vapor takes place (Frame 5). About 61 ms later after the ejected UDMH vapor and thin gellant layer burn, a luminous jetting flame can be seen on the right side of the droplet. After several eruptions, most of the UDMH is consumed and further fuel vapor eruption becomes weak (Frames 7 and 8). Because the experimental observations occurred within a space of 100 mm, the droplet cannot burn out, hitting the bottom surface of test chamber, and continues to burn. Experimental results (the hot plate combustion) are shown in the following subsection.

Figure 2. Picture series of falling and burning gelled propellant droplets.

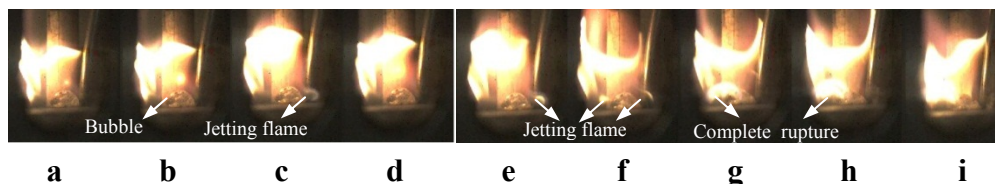


3.2. Gelled UDMH Droplet on Hot Plate

The droplet, which does not burn out during the free fall process, hits the bottom surface of the test chamber, and a luminous flame appears. As the UDMH is consumed, the increasing concentration of the gellant in the droplet surface creates an elastic layer, which prevents the majority of vapors in the inner layers of the droplet from escaping. As a result, these vapours are manifested in the form of bubbles formed near the contact area between the droplet and the bottom surface, primarily due to the increased conductive heat transfer rate. The ruptures in the flame structure shown in Figure 3c indicate the regions in the droplet surroundings where the vapour jetting occurs. Subsequently, the vigorous jetting flame formation in Figures 3d–f indicate that a large number of bubbles are formed inside the droplet during this time and the scale of the microexplosions is increased. After a certain period of time, the complete disruption of the flame structure in Figure 3g marks the point when the majority of

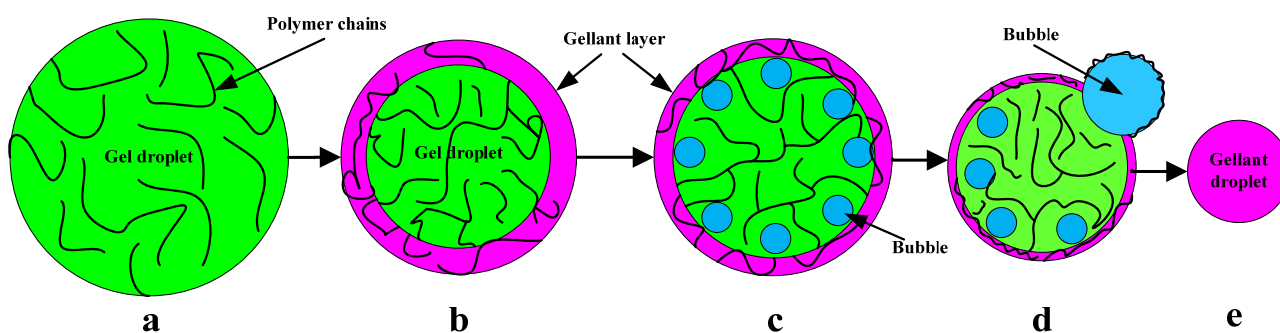
the vapors have been consumed or jetted out. The droplet now mainly contains the gellant and continues to regress like a normal single component fuel droplet.

Figure 3. Picture series of burning gelled propellant droplet on the hot plate.



Based on the description of the combustion of freely falling droplets and droplets on a hot plate, the combustion process of polymer gelled fuel can thus be divided chiefly into three stages: (1) Phase separation stage, namely a classical combustion stage (Figures 4a,b). This stage is characterized by a steady flame envelope formed surrounding the droplet, which is similar to the classical burning behavior of a single-component droplet. Within the droplet, the base fuel (UDMH) of the droplet surface is consumed; whilst the gellant layer begins to form gradually; (2) Bubble appearance, growth and vigorous microexplosions (Figures 4b–d). Microexplosions occur several times until most of the base fuel (UDMH) is consumed, which leads to no steady flame front around the droplet; (3) Gellant combustion stage (Figure 4e). The formation of fuel bubbles inside a droplet during the burning process has been reported by several authors when using slurry fuels or organic-based gelled fuels [4,8,9,14]. During the combustion of gelled UDMH several different interesting phenomena can be seen, such as bubbles being formed inside the droplet, expanding continuously and reaching almost the same order of magnitude as the droplets themselves. This is primarily due to the higher yield stress of polymer gellant than Thixotrol ST gellant and polymer chain structure.

Figure 4. Microexplosion mechanism of a gelled droplet.



3.3. Burning Rate

The influences of pressure and mixture ratio in the combustion chamber on the burning rate of droplets have been studied for liquid propellants. Natan *et al.* have conducted experimental studies on the combustion characteristics of hydrocarbon based gels (kerosene [4], JP-5 [6], JP-8 [10]), indicating that the amount of added gelling agent has a strong influence on the burning behavior of the droplets in comparison to the pure liquid. In this paper, the effects of pressure and oxygen fraction on the rate of burning were investigated by changing the pressure and the oxygen fraction in the test chamber.

Burning rate under different experimental conditions can be obtained and compared with a theoretically calculated burning rate constant.

The burning rate constants were calculated from the d^2 -law, given as [15]:

$$d^2 = d_0^2 - Kt \quad (1)$$

where d_0 and d are initial droplet diameter and instantaneous diameter respectively; and K is the burning rate constant.

The heat and mole transfer processes of the droplet combustion discussed so far are strongly influenced by external convection. The effects of external convection for a freely falling droplet on burning rate must be considered. According to the theory of droplet combustion under convection condition, a recommended relation for burning rate constant is:

$$K = \frac{d(d^2)}{dt} = \frac{4k_g Nu}{\rho_l c_{pg}} \ln(1 + B_T) \quad (2)$$

where B_T is the Spalding transfer rate, which is determined by the following equation.

$$B_T = \frac{c_{pg}(T_\infty - T_s) + \beta Y_o Q_R}{L_v + c_{pl}(T_s - T_0)} \quad (3)$$

where ρ_l is the gel density; c_{pl} is the gelled UDMH specific heat capacity; k_g is the thermal conductivity of ambient gas; c_{pg} is the specific heat capacity of ambient gas; T_∞ is the ambient gas temperature; T_s is the droplet surface temperature; T_0 is the droplet initial temperature; L_v is the latent heat of vaporization of UDMH; β is the stoichiometric ratio; Q_R is the heat of combustion; Y_o is the initial oxygen mole fraction and the Nu is the Nusselt number. The droplet falling velocity is calculated based on relative displacement and time interval of different images captured by fast-speed digital camera. Nu is found from the semiempirical correlations of Ranz ($Re = 10 \sim 500$):

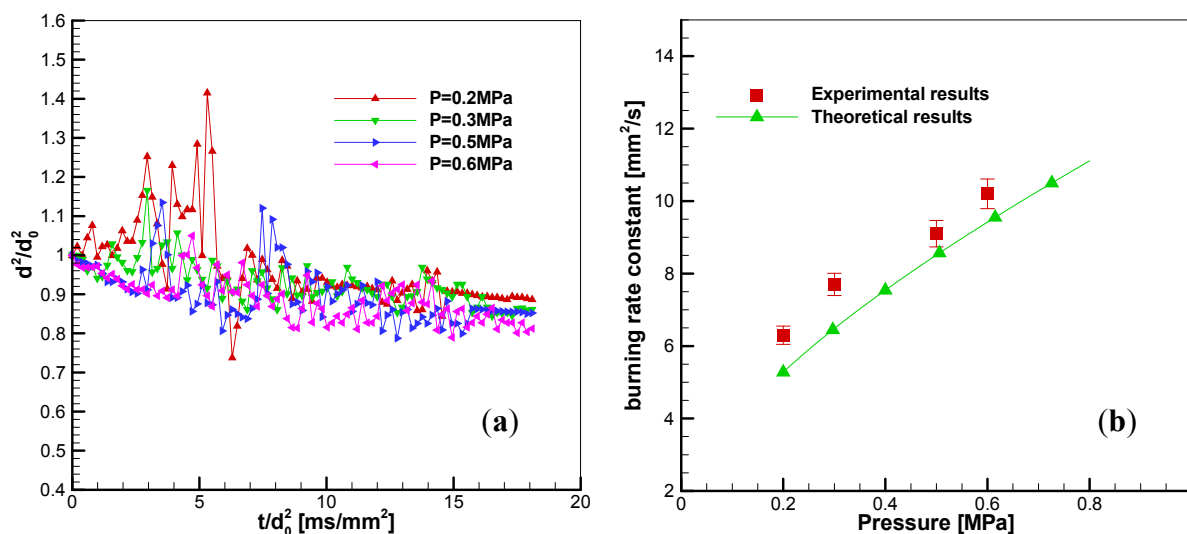
$$Nu = 2 + 0.6\sqrt{Pr}\sqrt{Re} \quad (4)$$

3.3.1. Effects of Pressure

In order to investigate the effects of pressure on the burning rate constant, four sets of experiments were conducted by pressurizing the test chamber from 0.2 to 0.6 MPa. The initial droplet diameter in these experiments was 2.26 ± 0.10 mm; initial temperature and oxygen fraction were 473 K, 0.5 respectively.

Figure 5a shows variations of normalized squared diameter versus the normalized time for different ambient pressure and constant oxygen fraction 0.5. It is noticeable that the peak number of normalized squared diameter and the peak value decrease with increasing ambient pressure. Besides, microexplosions appear later with higher pressure. These observations emphasize that bubble formation and probable microexplosions of the droplet are suppressed when the test chamber pressure increases. Similar results were observed by Mishra for ATF gelled propellant droplets [15].

Figure 5. (a) Temporal variation of droplet size for different ambient pressure at $Y_o = 0.5$; (b) Variation of burning rate constant with ambient pressure.



This happens because with increasing pressure, the boiling temperature of UDMH increases, heat up period becomes longer, resulting in microexplosions being delayed. Moreover increasing pressure can suppress the minor perturbations occurring due to bubble movements inside the droplet and weaken the intensity of microexplosions. In Figure 5b, a comparison between experimental results and theoretical results for variation of burning rate constant with ambient pressure is presented. With rising pressure, the burning rate increases. The reasons for this are two-fold. First, the oxygen concentration increases relatively near the droplet surface; the flame around the droplet is closer to the droplet surface, and this intensifies heat transfer. Second, rising pressure leads to the steady-state temperature of the surface layer increasing, and the latent heat of UDMH vaporization decreases. Solomon conducted experimental investigations on the combustion behavior of gelled MMH droplets, and pointed out that the burning rate has no apparent dependency on pressure, and increasing the pressures range of the experiment would eventually show a relation [20].

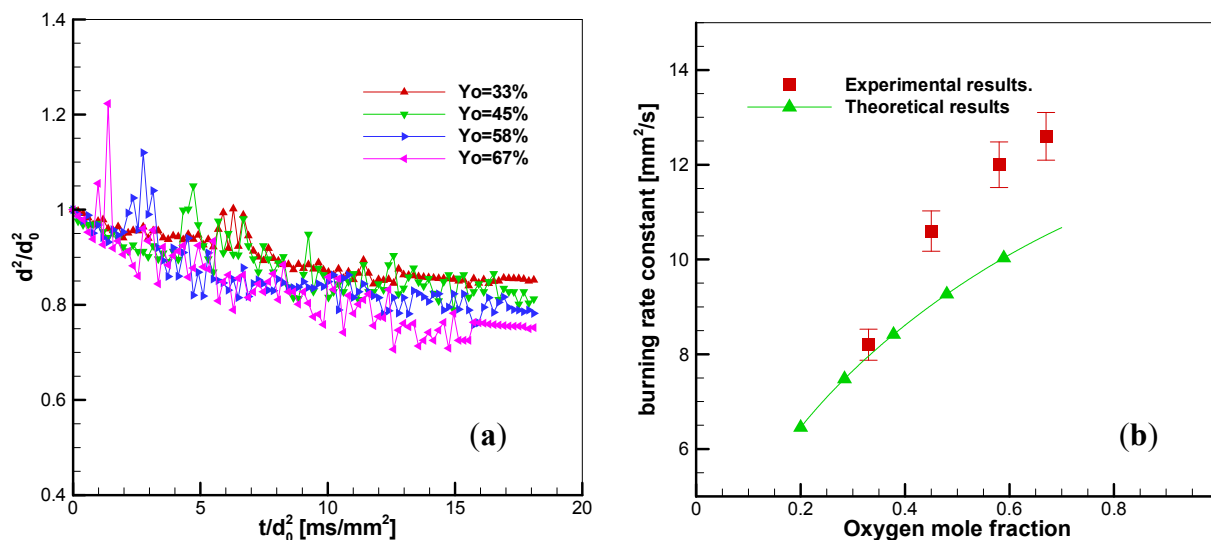
3.3.2. Effects of Oxygen Fraction

In combustion studies, oxygen fraction is one factor. In order to study the effect of oxygen fraction on the burning rate constant, four sets of experiments were conducted by changing the oxygen fraction of the test chamber from 0.33 to 0.67. The initial droplet diameter range studied in these experiments was 2.27 ± 0.1 mm; initial temperature and pressure were 573 K, 0.6 MPa respectively.

Figure 6a represents temporal variations of normalized squared diameter versus the normalized time for different oxygen fraction and constant pressure 0.6 MPa. With rising oxygen fraction, the number of peaks of normalized squared diameter and the value of peaks increases, moreover the microexplosion time is seen to decrease, which can be explained easily. Increasing of the oxygen fraction results in the rising of the flame temperature, and the additional heat arriving at the droplet surface is used for droplet heating, the bubble formation time decreases significantly, whilst pressure in the bubbles increases, which intensifies the microexplosions. Figure 6b depicts the variation of burning rate constant with oxygen fraction and comparison between experimental and theoretical

results. With rising oxygen fraction, the flame temperature increases, this intensifies the heat transfer, and induces an increase in the burning rate constant.

Figure 6. (a) Temporal variation of droplet size for different oxygen fraction at $P = 0.6$ MPa; (b) Variation of burning rate constant with oxygen fraction.



Figures 5b and 6b represent comparisons of the burning rate constant between experimental and theoretical results for different pressures and oxygen fractions, respectively. The reasons why the theory underestimates the burning rate constant arises from two aspects. On the one hand, freely falling droplets must be generated before the freely-falling droplet combustion experiment is conducted. The whole process including droplet detachment, ignition, burning and free fall cannot be tracked completely, especially for high viscosity gelled fuels. The lifetime of a droplet is calculated from droplet detachment and free falling, leading to a short droplet lifetime, which is also the disadvantage of the freely falling droplets combustion experiments. On the other hand, during the droplet combustion process, droplet deformation, bubble formation and growth, vapor jetting, leads to no steady flame front around the droplet, and intensifies the heat transfer. These factors are not taken into account for the theoretical prediction of burning rates.

4. Conclusions

In this paper, the burning characteristics of organic gallant-based gelled UDMH droplets is studied experimentally utilizing single-isolated free falling droplets instead of the conventional suspended droplet approach. Morphological transformations of the gelled UDMH droplet involved in the combustion processes are obtained by employing a fast-speed camera, whilst the effects of ambient pressure and oxygen fraction on burning rate are also analyzed. The results can be summarized as follows:

(1) The experimental observations indicate three distinct phases (gellant layer formation, bubble formation and growth, vapor jetting and microexplosion), and four main phenomena (droplet deformation, bubble formation and growth, vapor jetting and luminous jetting flame with “horn” shape) in the combustion process. Bubbles can reach almost the same order of magnitude as the

droplets. The high yield stress (compared with Thixotrol ST) and polymer chain structure of polymer gellant are put forward to explain the results.

(2) The burning rate constant is found to increase with both pressure and oxygen fraction. The microexplosion time is seen to increase as the pressure increases, while it decreases as the oxygen fraction increases. The intensity of microexplosions decreases with pressure due to increased resistance, and increases with the oxygen fraction due to increased flame temperature.

Acknowledgments

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