Comparison on Laser Ignition and Combustion Characteristics of Nano- and Micron- sized Aluminum

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Abstract

A comparative study was conducted on the ignition and combustion characteristics of nanometer aluminum (nano-Al) micron-sized aluminum (micro-Al) powder stacks. It is pointed out that the nano-Al and micro-Al stacks are significantly different in ignition mode by laser heating. In the static air flow with atmospheric temperature and pressure, ignition delay time of nano-Al is much smaller than that of micro-Al. The combustion of nano-Al is more intense and the self-maintenance performance is better than micro-Al. A model is built to analyze the combustion difference of nano-Al and micro-Al. It is presumed that the ignition and combustion characteristics depends on particle size, oxide film thickness and porosity of powder stacks.

1 Introduction

Aluminum is commonly utilized as a fuel ingredient in solid propellants to improve the combustion in rocket engines, because of its high reactivity and relatively large volumetric combustion enthalpy. An early study by Friedman and Macek[1-2] demonstrated that the burning time of 30-50μm aluminum is an exponential function to the diameter. For micro-Al particles, the energy release is insufficient due to their high volumetric heat capacity. Ignition temperature is upon melting of the oxide shell at 2350K[3]. In recent years, many researches have shown that the physicochemical properties of the nano-Al are quite favorable as compared with those of micro-Al counterparts. In a solid propellant, the addition of nano-Al fuel increases the burning rate by 70% compared to the same amount of ordinary micro-Al[4]. This is because that the fraction of atoms in the surface layer increases dramatically from 6 to 47% as the particle size decreases from 30 nm to 3 nm. The surface atoms have smaller coordination numbers and greater energies than the atoms in the interior regions of the particle[3]. Many works have been done to study the combustion properties of nano-Al, such as the melting temperature[5], ignition temperature, flame propagation speed [6], and burning time[7], etc. Regardless of the experimental results, it is inseparable from the mechanism behind them. There are current two mechanisms: diffusion oxidation mechanism (DOM) and melt-dispersion mechanism (MDM).

The preceding studies have been carried out at different scales to improve the thermites of the Al. However, most of researches focused on aluminum-containing composite materials[8]. The complexity of

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composites made it difficult to analyze the thermite mechanism and unveiled the interaction between aluminum particles and others. The combustion mechanism of pure Al is of great significance for studying its additive performance. Therefore, the objective of this study is to compare the ignition and combustion characteristic of pure nanometer- and micrometer- scale Al by laser heating. Six samples with different sizes were examined, in average diameters of 56.0nm, 74.4nm, 93.4nm for nano-Al and 2.9μm, 6.1μm, 10.8μm for micro-Al. Ignition and combustion process were recorded by high-speed camera and infrared thermal imager, and the characteristic differences between nano-Al and micro-Al were analyzed and discussed. Finally, a model was built to explain the ignition and combustion behaviors related to the particle size, oxide film and porosity of powder stacks.

2 Experimental methods and materials

The schematic of experimental setup is shown in Fig.1. A synchronous trigger controlled the high-speed camera, infrared thermal imager, silicon photodetectors and PMT. The laser beam was split into two beams. One beam was shaped by beam expander, reflector and lens to ignite the samples. The other beam directly entered into the silicon photodetector to record the response of laser. The optical fiber probe connected with PMT is placed on one side of the sample parallel to the burner, collecting radiation signals. The high-speed camera shot the burner at an angle of 35 degrees, while the thermal imager shot at 75 degrees. The laser (λ=1064nm) was operated in continuous wave mode and produced a maximum power density of approximately 99.98 W/cm^2. Its electro-optical response was measured to be 50μs. Ignition delay time was determined as the difference between laser onset and the first visible light output recorded by the high-speed camera at 11,000 fps.

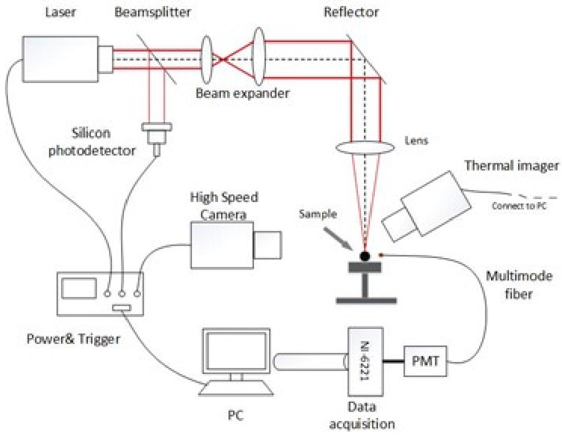

Fig. 1 Schematic of experimental setup

The nano-Al powders (the oxide film δ≈3nm) were purchased from Changsha Tianjiu Corporation and the micro-Al (δ≈2-8nm) were purchased from Beijing Deke Daojin Corporation. All experiments on samples combustion were carried out in air at the room temperature of ~23℃, the pressure of 1 atm and a relative humidity of ~55%. Al powder were piled up naturally on the burner flat.

3 Results and discussion

3.1 Ignition delay and temperature rise

Ignition delay time was recorded five to eight times for each Al sample. Figure 2 shows that ignition delay time is a function of particle size. Ignition delay time of 56.0, 74.4 and 93.4 nm are 2.08, 1.84 and 1.81ms. Ignition delay times of 2.9, 6.1 and 10.8μm are 3.56, 5.87 and 8.03ms. It can be clearly seen that ignition delay time of nano-Al decreases with increasing particle size under the same ignition and operational conditions, while ignition delay time of micro-Al increases with increasing particle size. Obviously, ignition delay time of micro-Al samples is overall greater than that of nano-Al samples. There might be a minimum value (critical point) in sub-micron size between 93.4nm to 2.9μm. Bockmon et al.[9] demonstrated similar experimental results that the burning rate of nano-Al powder has a critical value between 80-110 nm as a function of particle size. It indicates that ignition delay time is dependent of Al particle size.

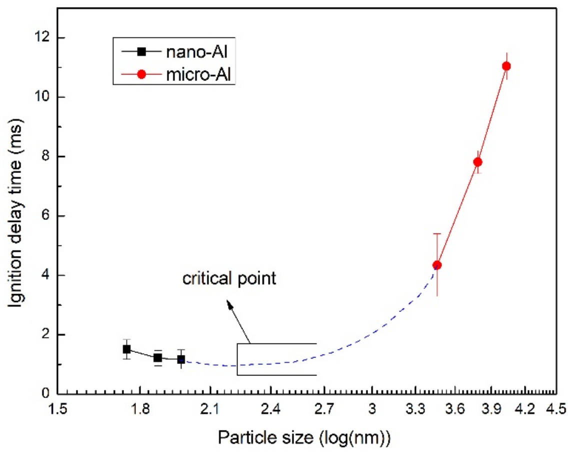

Fig. 2 Ignition delay time of six samples

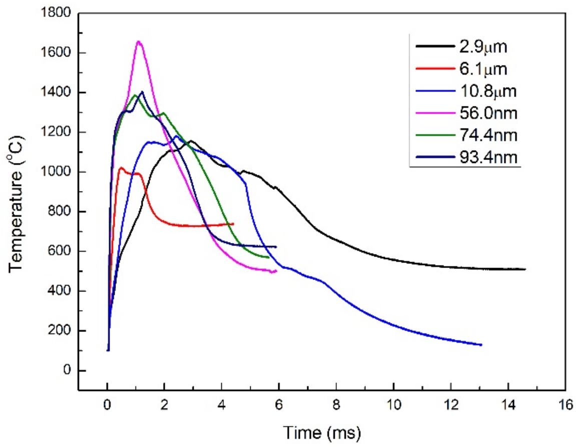

Fig. 3 Temperature rise of six samples

Fig. 3 shows the temperature curves of Al powder stacks recorded by infrared thermal imager (200 fps). The temperature curves of three nano-Al samples are steeper than those of micro-Al, indicating that the temperature rise rate of nano-Al is larger. However, the heating rates and the maximum temperature do not monotonously decrease with increasing particle size. It might be that the temperature rise rates of Al powder stacks are not only affected by particle size, but also depend on oxide film thickness and porosity of powder stack, resulting in different ignition and combustion characteristics.

3.2 Combustion and flame

The same mass of aluminum powder (about 2.5mg) were naturally stacked on the burner flat (see Fig.4 a1 and b1). The laser was operated in its original state of a Gaussian beam distribution, special care was taken to ensure that the beam was aligned with the sample center. The parameter *t* in the pictures represents the synchronization time of the two cameras. In the preheating stage, the sample temperature rose quickly when the laser irradiated on the samples (See Fig.5). In the second stage, the time taken for the nano-Al to reach the maximum temperature was 800 ms (c5), while the time at which the micro-Al reached the maximum temperature was 2375 ms (c4). It shows that the heating rate of nano-Al is much faster than that of micro-Al. When the t=1.73 ms (while the t=12.55ms in b2), Al sample appeared the first visible light (a2) and the ignition happened at this moment. After ignition, the combustion waves propagated through the sample surface (a2-a4 and b2-b4). The energy from the continuous laser and the energy released by burning aluminum made the temperature rise and the area of the brighter light enlarged (a4-a5 and b4-b5). Since the sample mass was tiny, we did not observe a significant flame front. After a while, the samples were cooled down quickly to extinguish.

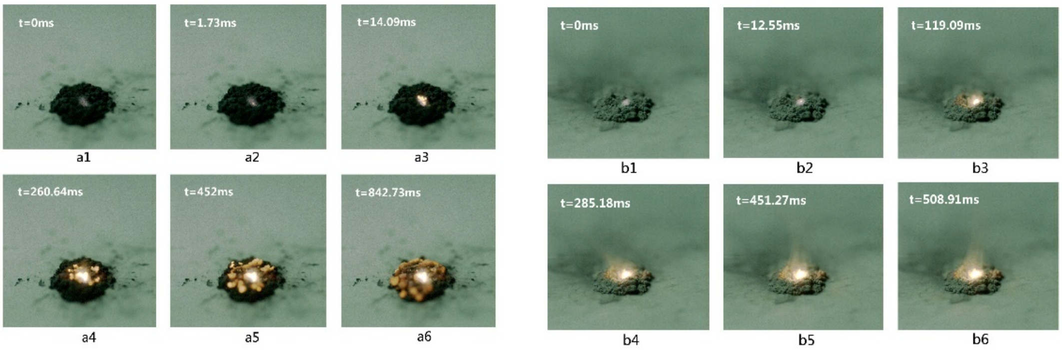

Fig. 4 Burning sequence of the Al powder (a1~a6 for 93.4nm and b1~b6 for 10.8μm).

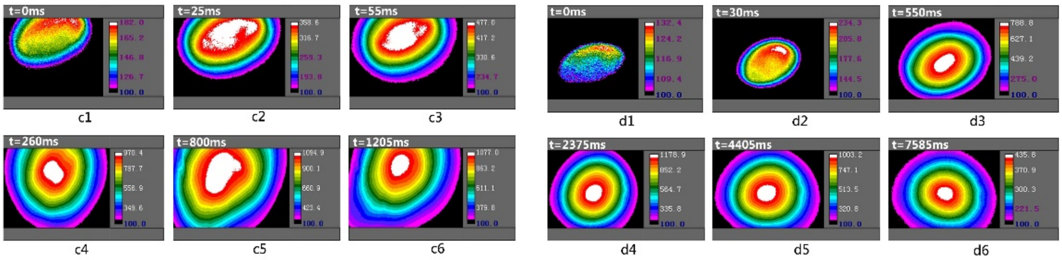

Fig. 5 Temperature distribution (c1~c6 for 93.4nm and d1~d6 for 10.8μm).

Comparing the combustion process of nano-Al and micro-Al, we find that nano-Al reacted more rapidly and completely than micro-Al under the same conditions. The nano-Al particle group can maintain combustion by the heat released from itself, while the flame of micro-Al cannot diffuse outside the laser spot. This is because the melting temperature of nano-Al is much lower than that of micro-Al[6].

3.3 Combustion model and analysis

A model is built for analyzing laser ignition and combustion difference of nano-Al and micro-Al powder stacks, as shown in Fig.6. The model demonstrates that the ignition and combustion characteristics of aluminum powder depends on the particle size, oxide film thickness and porosity.

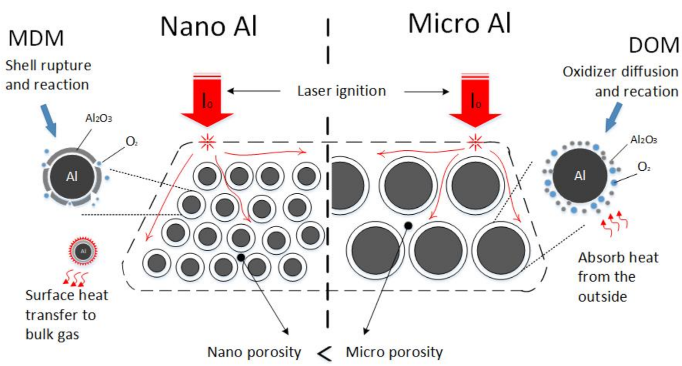

Fig. 6 Combustion model for nano-Al and micro-Al.

Under the same laser power and operational conditions, the combustion of nano-Al powder stack is mainly controlled by MDM, while the micro-Al is mainly controlled by DOM. For the MDM control mechanism, the key parameter is the ratio of Al core radius R to shell thickness δ, M=R/δ[14]. The M is the critical value of the flame propagation speed of aluminum as a function of particle size. Our experimental result shown in Fig.2 demonstrated similar performance. For the DOM control mechanism, the flame propagation speed of micro-Al increases with the increase of surface area to volume ratio and oxide film thickness. Thus, the ignition delay time of micro-Al increases with increasing particle size, which is agreement with the results in Ref.[16].

The thermal conductivity of Al_2O_3 is not negligible during ignition. For nano-Al, the effect of the oxide film will be greater than that of micro-Al. The increase in the proportion of oxide film reduces the heat absorption of nano-Al. Therefore, nano-Al particles will make it more difficult to complete the cracking of the film. As the particle size of micro-Al increases, the thermal insulation effect of the aluminum oxide significantly reduces. Micro-Al have greater endothermic efficiency and heat loss rate. The heat absorbed by micro-Al only accelerates the progress of DOM and does not completely change its oxidation mechanism.

The combustion of aluminum powder heated by a high-power density laser can be divided into two stages: ignition and combustion. During ignition, the aluminum powder is primarily heated by the laser and the heat transfers from the surface. In the viewpoint of energy balance, the evolution of the aluminum particle temperature (*T*) can be given by:

$$\frac{1}{\kappa}\frac{\partial T}{\partial t} - \nabla^2 T = Q_S + Q_L + Q_{HL} \tag{1}$$

This equation expresses the evolution of the particle temperature caused by the heat generated by the exothermic chemical reaction (Q_S), the volumetric heating caused by laser (Q_L) and the heat loss of particle stack ignition (Q_{HL}). The energy terms of Eq (1) can be expressed as following:

$$Q_S = \frac{\rho Q K}{\lambda} e^{-E_a/RT} \tag{2}$$

$$Q_L = I_0(t)\frac{\alpha}{\lambda} e^{-r^2/\varpi^2} e^{-\alpha_z} \tag{3}$$

$$Q_{HL} = hA(T - T_{\text{ex}})(1+\eta)^{-1} \tag{4}$$

Where *h* is the heat transfer coefficient of the sample, *A* is the sample surface area, *T* is the temperature of the sample and T_{ex} is the external gas temperature.

It is assumed that the overall physical properties (λ, ρ, c, E_a) of the substance during ignition are unchanged. Since the laser power and area are constant, the volumetric heating caused by laser (Q_L) can be simplified as a function of the absorption coefficient α of the particle stack to the sample. The effect of the heat loss on the laser ignition is considered in Eq. (1) and the simplified model is expressed in Eq (4). Assuming the temperature required for ignition of the sample particle stack is T_{ign}, the ignition delay time t_{ign} required for the particle pile to reach T_{ign} under laser action is mainly controlled by Q_S, Q_L and Q_{HL}.

As shown in Fig. 3, the heating rate of nano-Al in the experiment is larger than that of micron-Al. In Eq (2), the nano-Al is produced at a much faster rate than the micro-Al due to its lower melting temperature and faster reaction rate. As a result, the nano-Al ignition delay time is much smaller than that of micro-Al. However, in the nanometer scale, the nano-Al decreases its heat release rate faster than the heat loss as the particle size decreases. This causes the ignition delay time of the nano-Al to decrease as the particle size becomes larger (left half of the curve in Fig. 2). For micro-Al, the larger particle diameter makes the η value larger [18], and the heat transfer is more pronounced in the ignition of micro-Al. And as the particle size increases, the rate of change of the Q_L term is greater than the rate of Q_S. Therefore, the ignition delay time of micro-Al increases as the particle size increases (right half of the curve in Fig. 2). Another factor of porosity cannot be ignored in the sample powder stack. There are large differences between nano-Al and micro-Al in density, specific heat and porosity. The size of porosity will affect the magnitude of c, α and Q_L. The micro-Al powder stack has a bigger porosity and air content, which allows the powder stack to absorb less heat. As a result of the interaction of the various parameters, the micro-Al powder stack is more difficult to ignite than the nano-Al.

4 Conclusions

Ignition delay time of nano-Al is much smaller than that of micro-Al. Ignition delay time of nano-Al decreases with increasing particle size, while micro-Al increased. There might be a critical value at the sub-micron level to minimizes ignition delay time. Nano-Al burns more intensely than micro-Al and has a shorter burning time. The flame of nano-Al can diffuse naturally, while micro-Al does not. Through model analysis, the differences in ignition between nano-Al and micro-Al powder stacks result from the combination of oxidation mechanism, particle size, oxide film thickness and porosity.

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