Oscillatory combustion characteristics of micron-size aluminum powder in sound field

Junwei Li, Yanbin Wang,Ningfei Wang

School of Aerospace Engineering, Beijing Institute of Technology,

Beijing 100081, PR China

1 Introduction

Aluminum particles have been used for a long period of time as an energetic additive in propellants and explosives. Al has high volumetric heat when compared to other metallic additives and still performs fairly well on a mass basis without some of the other practical limitations, like toxicity or expense, of the other additives. For these reasons aluminum is a very effective way to add a great deal of heat to a combustion system with the least amount of weight[1]. Additionally, in rocket motor propulsion systems the Al particles act as an effective damping mechanism to suppress combustion instabilities.

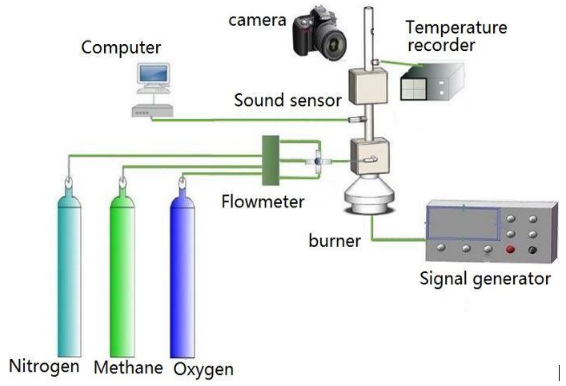
Combustion instability in solid propellant rockets has been studied intensely for over four decades but complete understanding remains elusive. By adding addition fine metal particles to viscously damp acoustic oscillations has been One solution to the stability problem[2,3]. Although combustion of aluminum particles and their cohesive oxidation products will bring about loss of viscous damping acoustic oscillations, but combustion of metal particles to release energy can affect sound field of combustion chamber, and even enhance acoustic oscillation. Thus using particles as acoustic suppressants demands intimate understanding of the interactions between particles and the motor's acoustic field.

2 Experimental setup

The experimental apparatus, as shown in Fig. 1, mainly consists of gas supply system, acoustic excitation and data acquisition system, aluminum supply system and a flat-flame burner. Flow rates of three gases (methane, nitrogen and oxygen) are controlled by mass flow controllers (D08-2F, range 500mL, 5L and 2L). Acoustic wave is generated by a signal generator (Model XFD-8c) and a speaker (3Ω, 4W), and recorded by sound pressure sensor and an acquisition card. Gas temperature is measured by a thermocouple (S-type, diameter 0.5mm). Aluminum mass is measured by an electronic balance (accuracy 0.1mg). As shown in Fig. 2, the flat flame burner is composed of quartz glass tube (inner diameter 10mm) and porous media (foam nickel).

The aluminum powder is manufactured by Shanghai Naiou Nanotechnology Co.,Ltd and has average particle sizes of 10μm, 20μm and 30μm. The aluminum purity is about 99.9% and molecular mass of aluminum is 26.98 g / mol.

Correspondence to: david78lee@gmail.com



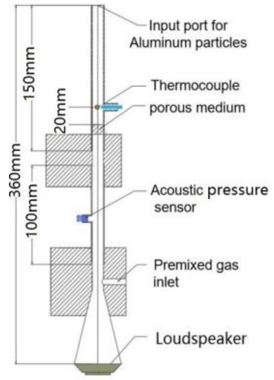


Fig. 1 Schematic of the experimental setup

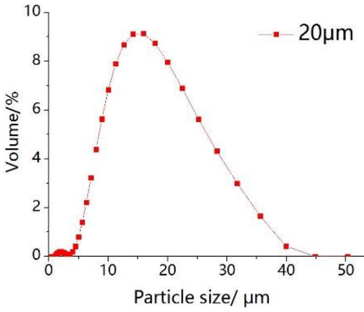
Fig. 2 Methane premixed flat-flame burner

The experimental procedure is as follows. Firstly, flow rates of three gases are adjusted to desired value. The mixture is ignited by an electric spark at the burner outlet. The flame moves into the quartz tube, and is stabilized on the surface of the porous medium. Then, the signal generator and loudspeaker are turned on to the desired value. After weighing with an electronic balance, 1.1 mg of Al particles are taken with a Paper funnel and the weighed aluminum powder is injected into the burner from the outlet. When the aluminum particles fall toward the flame, they are gradually heated, and then burns at the flame surface, emitting bright white light. At this stage, the injection of aluminum powder in the experiment is a one-time injection, not a continuous injection process. At present, we are designing a system composed of stepper motors to achieve a continuous injection of a small amount of aluminum powder process. Steady gas temperature above the flame is measured by a S-type thermocouple placed 20 mm above the porous medium. The aluminum flame is recorded by a JVC video recorder (Model GC-PX100). Sound pressure in the burner is measured by a sound pressure sensor (BSWA MPA416) and recorded by NI 9234 data acquisition card. Sampling frequency of the card is 50KHz and the sampling time lasts 3 second. In this paper, combustion of 20μm aluminum particles is firstly studied, and then combustion of aluminum powder with different sizes and different masses are analyzed.

3 Results and discussion

3.1 Analysis of 20μm Aluminum Combustion

Experiments of Al particles combustion are conducted in the flat-flame burner under room temperature and atmospheric pressure. Al particles of 1.1 mg 20μm are injected into the burner. Low-frequency acoustic oscillation of sine wave is produced by the signal generator and applied on the burner. Frequency of acoustic wave is 200Hz and amplitude is 0.097Pa. Flow rate of the mixed gas is 1.46L / min and the equivalent ratio is 0.9. The gas flame temperature is 1093 K, lower than the adiabatic flame temperature 2373 K. Particle size distribution of the Al powder is shown in Figure 3, measured by a granulometer (Hydro 2000MU). This figure shows that the most of Particles are in the range of 12~ 30μm. In the experiment, when 1.1 mg of aluminum particles were injected into the burner at one time, the combustion process of aluminum particles was recorded by VCR. According to the sound pressure curve in Fig. 5(a), the images of aluminum powder burning at different times were intercepted to describe the process of aluminum powder combustion. Photos of Aluminum combustion are shown in Fig4. It can be seen that at t = 0.804s, the aluminum particles are injected into the burner and heated by hot surrounding gas. The flat flame is not disturbed by the Al particles. When the aluminum particles reach the flame zone, the particles are over melting point of pure aluminum.Pure aluminum in the core is melt and becomes liquid. Since expansion coefficient of aluminum oxide is less than that of aluminum(aluminum oxide: 23.2×10^{-6}/K, aluminum:8.4×10^{-6}/K). The aluminum oxide shell is ruptured by the liqiud aluminum, and the liquid aluminum in the core is squeezed out from the crevasse. Finer droplets of aluminum react with the surrounding gas. At t = 1.117s, the intense burning can be seen from Fig4. Then, the surface of the formation of new oxide layer prevents the reaction of aluminum droplets. At t = 1.239 ~ 1.280s, the aluminum combustion is finished.



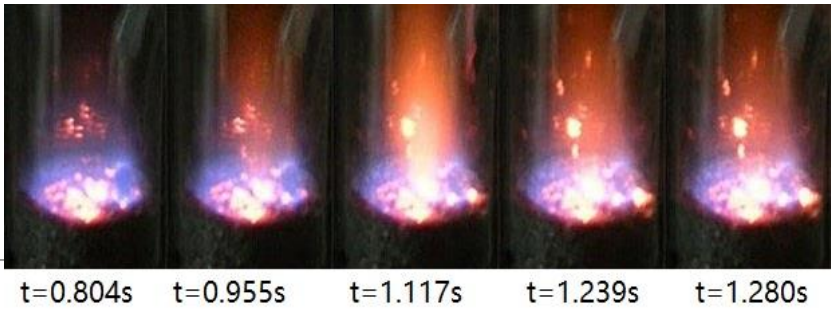


Fig. 3 Particle size distribution for aluminum powder with nominal diameter of 20μm

Fig. 4 Photos of Aluminum combustion at different time

Figure 5(a) shows variation of sound pressure during combustion of aluminum powder. From figure 5(a), it can be seen that combustion of aluminum particles can be divided into three stages: preheating stage, burning stage and damping stage. At the preheating stage, t0 ~ t1,(t0=1.0955s,t1=1.1112s), aluminum particles are preheated by the flame. Sound pressure decreases from -0.018Pa to -0.82Pa. This may be due to the absorption of aluminum particles. In the preheating stage, as it can be seen from Figure 5 (b), there is no high-frequency signal, indicating that the Al has not started to burn.At the burning stage, t2~t3(t2=11236s, t3=1.11336s), the burning time is very short, only 1 ms. This is due to that the aluminum oxide shell breaks and the liquid aluminum is squeezed out of the shell to form finer droplets of aluminum. Aluminum droplets react with surrounding air. The pressure sharply increases from -0.82Pa to 2.47Pa, and high-frequency oscillation is induced. Temperature increase rate of Al particles ν1 is calculated by T-T0 / δ1 (T = 1093K, ambient temperature; T0=293K, room temperature; δ1 = 0.0157s, δ1 = t1-t0), ν1 =50955K / s. The value of ν1 affects the rate of oxide layer breakage of aluminum. Under the heating, aluminum oxide crystals will morphological changes, but will not rupture. Pressure increasing rate ν2 is expressed by δP / δ2 (δP = 3.29Pa, δP = P2-P1; δ2 = 0.9ms, δ2 = t3-t2), ν2=3655Pa/s. The value of ν2 affects the rate at which liquid aluminum is extruded from the hard shell and the degree of high-frequency acoustic oscillations induced. From equation p '= P-P0, P is the measured pressure, P0 is the averaged sound pressure intensity of 200 data points, p' is the dynamic sound pressure. The dynamic sound pressure variation with time is plotted in Fig.5(b). After t3, the sound pressure curve oscillates, which is shown that combustion of aluminum particles is finished. Amplitude of pressure oscillation is decreasing due to damping effect of the solid combustion products.

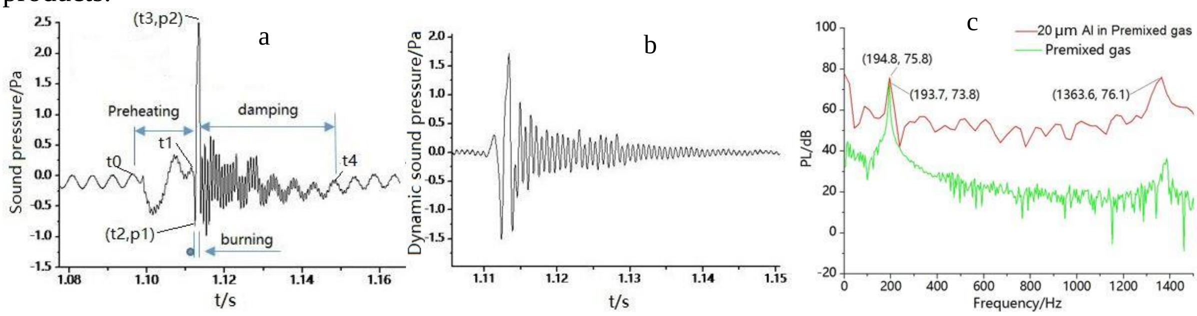


Fig. 5 Sound pressure of aluminum powder combustion. (a) Sound Pressure history, (b) Dynamic soun d pressure, (c) Sound pressure level - frequency curve

Sound pressure before and after the aluminum powder combustion is shown in Fig. 5 (c). Before injection of aluminum powder, frequency of the forced oscillation is 200Hz. When only gas mixture is burned in the burner, the measured sound pressure is 73.8dB. PL represents the sound pressure level. It can be found that after the aluminum powder is added into the burner, high-frequency oscillation(1363Hz) is measured. PL of the high-frequency oscillation is 76.1dB in the same order of the low-frequency oscillation. In this case, the equivalence ratio is 0.9. According to the thermodynamic calculation, the specific heat ratio of the combustion products k = 1.22 and the gas constant R = 286 J / kg · K can be obtained. According to the equation $c=\sqrt{kRT}$, the average sound speed 618m / s. By thermoacoustic frequency equation can be expressed as:

$$f = \frac{nv}{4(L + 0.4d)}$$

Where v is sound speed, L is whole length of the burner, d is diameter of the burner, f is the reasonate sound frequency, "n" is an odd number (1,3,5……). The burner length L = 360mm, calculated inherent acoustic vibration frequency of the burner is 1273Hz, close to the high-frequency oscillation 1363.6Hz. Therefore, the peak around 1363Hz is interpreted as the signature of a 3/4 wave mode of the burner.

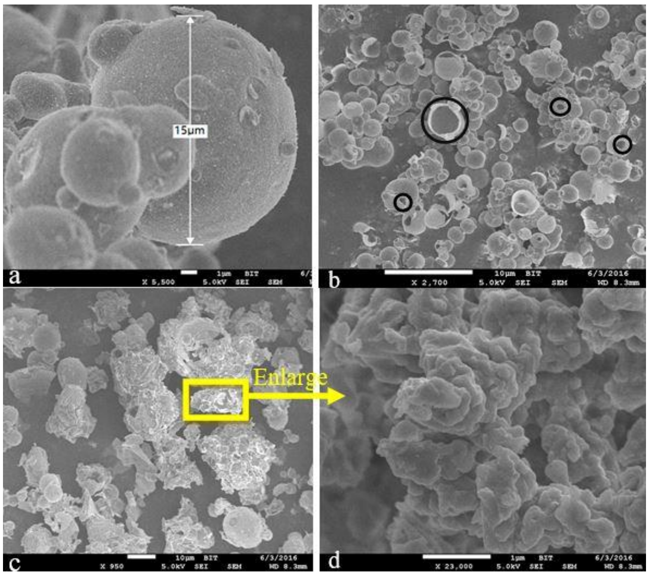


Fig 6 SEM photos of aluminum particles. (a) aluminium powder before burning, (b) rupture of aluminum oxide, (c) agglomeration of combustion products, (d) magnification of agglomeration of combustion product

Combustion products of aluminum is scanned using SEM as shown in Fig6. Aluminum powder before burning is shown in Figure 6 (a).It can be seen that one aluminum particle is spherical and diameter is about 15μm. In Fig. 6 (b), as shown by the round mark, irregular polygonal cleavages appear on the surface of most of the combustion products. The ratio of crack diameter to particle diameter is about 0.25. The rapture of aluminum oxide is due to that the expansion coefficient of aluminum oxide is less than that of aluminum. When the aluminum particles are heated rapidly, the Al_2O_3 crystals can not be converted into other forms of crystals. During fast heating and, consequently, loading, such stresses do not have time to relax and cause the dynamic fracture and spallation of the alumina shell[4]. In Fig. 6 (c), When the solid combustion products are heated for a prolonged period of time, solid products in the original cracks continue to pile up. These products are formed by agglomeration of extruded liquid aluminum. It is no longer spherical, and stacked sheet layers are formed, as shown in Fig. 6 (d).

3.2 Effects of Particle Size of Aluminum Powder on Combustion

In this study, three sizes of aluminum particles of 10μm, 20μm,30μm are tested. The mass of each aluminum powder is 1.1 mg, and injected into the burner. External sound field and combustion conditions are Same as above. The frequency spectrums of sound pressure are shown in Fig.7. It can be seen that there are 200Hz low-frequency oscillations caused by the external acoustic field. Amplitude of the low-frequency increases from 0.08Pa to 0.16Pa with the increase of particle size. In addition, Figure 7 also shows that all particles with different size to stimulate high-frequency oscillations. With the increase of particle size, intensity of high-frequency oscillation increases.

Sound pressure intensity for three particle sizes are shown in Figure 8. With increase of particle size, PL of low-frequency oscillation increases from 73dB to 77dB, and PL of high-frequency oscillation increases from 54dB to 67dB. This indicates that with increase of particle size, the intensity of high-frequency oscillation is stronger. This is because the thickness of the alumina shell increases with the particle size, and the pressure of the molten aluminum core increases.

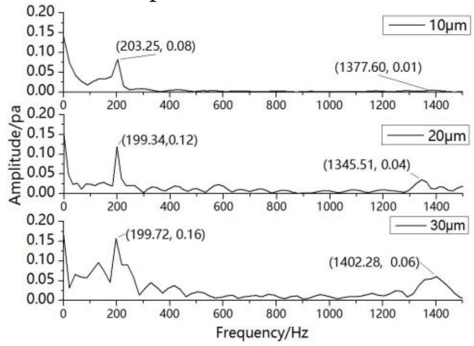


Fig. 7 curve of oscillation Amplitude - Frequency

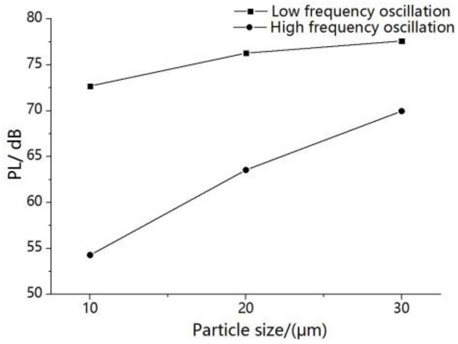


Fig. 8 Pressure level variations with particle size

Dynamic sound pressure of aluminum combustion is shown in Fig 9. As time increases, the pressure oscillations decay. This is due to that the solid combustion products damp sound oscillation. The sound pressure can be expressed by exponential relationship $p' = p_0' e^{\alpha t}$, where p_0' is the maximum sound pressure, and α is defined as amplitude decay constant. The decaying constants for different particle size can be calculated, and are -216, -120, -60.6Pa / s respectively. With the increase of aluminum particle size, the decaying constant decreases, which is consistent with the conclusion that smaller particles have strong attenuation ability to high-frequency oscillation[5] .

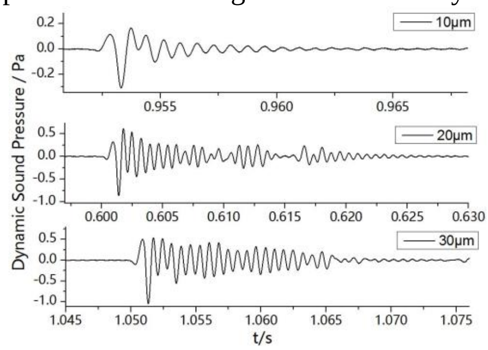


Fig. 9 Dynamic sound pressure variation with time

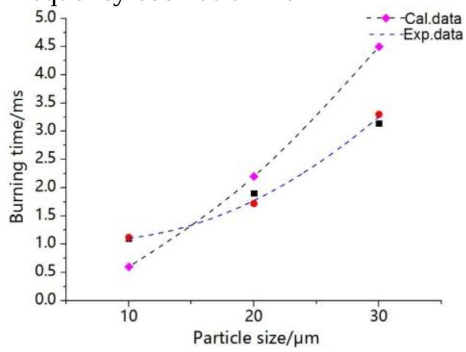


Fig. 10 combustion time variation with particle size

In order to describe the combustion process of aluminum particles, a lot of researches have been done. Beckstead[6] put forward a representative D^nmodel through a large number of experiments, and the suggested range of diameter index is 1.5-1. 8. Besides, a large number of studies have shown that the best results are obtained when n is 1.8. Burning time can be calculated by the equation:

$$t_b = \frac{c D_p^{1.8}}{X_{eff} P^{0.1} T_0^{0.2}}$$

where X_{eff} is the effective oxidizer concentration, $X_{eff} = C(O_2) + 0.6\, C(H_2O) + 0.\ 22C(CO_2)$, p the pressure in atm, T0 the initial temperature in Kelvin, Dp the particle diameter in μm, and c a constant ($c=7.35\times10^{-6}$). The burning time is quadratically proportional to particle size and is weakly dependent on the temperature and pressure of the gas. These suggest that the burning rate of Al particle is controlled by mass diffusion phenomena. According to the D^n model, combustion time of aluminum powder is calculated. As shown in Fig. 10, the calculated values agree well with the measured value. The measured time is defined as t1-t2, as shown in Fig5(a), which is plotted in Fig10. With the increase of the particle size, burning time increases from 1 to 3ms.

Table 1 Measured values of different particle size

D/μm	（t1-t2）/ms	f1/Hz	P1/Pa	f2/Hz	P2/Pa	α/(Pa/s)
10	1.11	200	0.09	1381	0.01	-200
20	1.81	198	0.15	1352	0.05	-132
30	3.22	199	0.20	1399	0.07	-70

f1 average frequency of low-frequency oscillation, P1 low-frequency amplitude of oscillation, f2 average frequency of high-frequency oscillation, P2 high-frequency amplitude of oscillation, α decaying constant

The measured values for different particles are shown in Table 1. When particle size is 30μm, the combustion time is the longest, and corresponding pressure amplitude of low-frequency and high-frequency oscillations are the largest. In Table 1, the frequency of low-frequency oscillation is 200Hz, which is unchanged for three particle sizes, the frequency of high-frequency oscillation in the range of 1370-1410Hz, This indicates that change of particle size has little effect on the oscillation frequency. Frequency of the high-frequency oscillation depends on the burner length.

4 Conclusions

Experimental studies were carried out to investigate ignition and combustion characteristics of aluminum particles. The following conclusions can be drawn:

(1) Aluminum combustion in the burner can stimulate high-frequency oscillation, and the sound pressure of high-frequency oscillation and low-frequency oscillation are in the same order of magnitude. When the aluminum particles are rapidly heated (Temperature increase rate v1 =50955K /s), the internal melting and expansion of aluminum particles cause the rupture of the oxide layer.

(2) With the increase of aluminum particle size, the damping coefficient decreases, burning time of the aluminum particles increases from 1 to 3ms. As the mass of aluminum powder increases, the pressure increasing rate increases dramatically from 800 to 1.1×10^4 Pa/s. Peak value of the high-frequency pressure increases from 55dB to75dB with More intense sound waves excited out.

(3) Combustion of micron aluminum powder can be divided into three stages: preheating, burning and attenuation. At the preheating stage, aluminum particles are preheated by the flame. At the burning stage, aluminum oxide shell breaks and the liquid aluminum is squeezed out of the shell. In the moment of alumina hard shell rupture, sound pressure increases rapidly. At the attenuation stage, aluminum droplets react with the surrounding oxygen, and stacked sheet layers are formed.

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