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Studies on the burning of micro- and nano-aluminum particle clouds

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Abstract

The aim of the present work is to reduce the burning time of aluminum particles with the ultimate goal to improve the performances of solid propellants. Aluminium nanoparticles have gained importance because of their increased reactivity as compared with traditional micro-sized particle. Decreasing the size of Al particles increases their specific surface area, and hence decreases the burning time of the same mass of particles. Nevertheless another consequence of decreasing the particle size is an increase of alumina mass fraction in the reactant powders passivated in air. An experimental program is initiated to determine flame propagation velocities of micro-sized (around 6 µm) and nano-sized (around 250 nm) aluminum particle clouds. Another goal of this study is to estimate the gas phase temperature from AlO molecular spectra and the temperature of condensed phase emitters in the flame using emission spectroscopy. To this end, an experimental set-up is developed to investigate the flame characteristics of particle clouds ignited by an electric spark in a glass tube.

The present results show that nano-sized Al particle clouds burn faster than micro-sized particle clouds for the same global particle mass concentration in air. The cloud flame propagation velocity depends also on the particle concentration. The temperature measurements indicate a consistent value around 2900 K for all nano-Al particle burning clouds and 3300 K for micro-Al particle clouds. The results of the condensed phase temperature show, first a stable temperature and then a decreasing trend along the axis of the flame.

Introduction

The available literature shows that Al nanopowder can significantly improve the combustion properties of some energetic materials, especially for propulsion applications. Solid propellants containing Alex nanopowder exhibit burning rates much higher (in some cases as much as 5 to 20 times higher) than the propellant formulations containing regular Al powder [1]. Further, HTPB-based (HTPB: hydroxyl-terminated polybutadiene) solid-fuel formulations containing Alex or other nanopowders demonstrated mass burning rates in a hybrid rocket up to 50% higher than the baseline formulations with regular Al powder [2]. These superior performances of the energetic compositions containing Al nanoparticles compared to the conventional micro-sized particles are related to higher specific area and reactivity of nanopowders.

In the present work, the investigation is related to combustion studies of aluminum particles injected into a tube and ignited by an electric spark after the formation of a cloud. The experiments are conducted for two purposes: first to measure the flame front propagation velocities for different sizes of the Al particles, second to measure the temperature of the gas and the condensed phases during cloud combustion.

Experimental setup and methods

Figure 1 shows the experimental setup used for studying the combustion of nanoparticle clouds in air under normal conditions of pressure and temperature. The combustion chamber is essentially a quartz tube (inner diameter of 14 mm, height of 180 mm giving an inner volume of 27,7 cm^3), whose bottom end is closed by a porous metal plate, and the top end is closed by a thin cellulose filter. For the given reactor volume, the stoichiometric conditions in air correspond to 9.3 mg of Al mass. The present experiments are performed for different conditions corresponding to a global Al concentration from 200 to 2000 g m^{-3}.

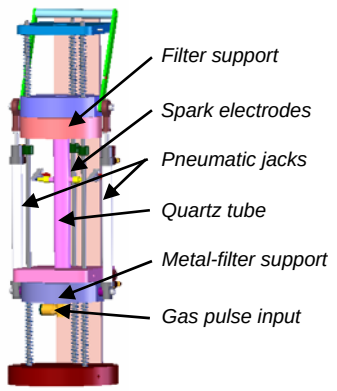

Figure 1: Combustion apparatus.

The powder sample is placed in the bottom of the tube. Injection of particles and formation of the cloud inside the tube are achieved by a pulse air injection, using an electrical valve, to carry and disperse the

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particles in the tube. The injection pressure and duration are adjusted to achieve an optimum distribution of the powder inside the tube. After suspension, the cloud is stationary due to low sedimentation velocities of Al nanoparticles within the range of 10^{-5}-10^{-3} cm/s. The particle cloud is ignited by a spark placed on the axis at 30 mm from the top section of the tube. In these experiments, this ignition section was selected as providing the optimum ignition probability for all the combinations between the injected powder quantity, the injection time and pressure. A high-speed video camera (Phantom V5.0 with 1000 fps at a maximum resolution of 1024 x 1024 pixels, or higher frequency by reducing the resolution) is used for the flame front observations and propagation velocity measurements. The propagation velocity of the burning cloud is determined for different Al nanopowder samples by image analysis.

For the temperature measurements, particle cloud flame emission spectra are recorded using a high resolution spectrometer in the visible range. The fitting of bands Δv = –1, 0, +1 of the AlO emission is used for the determination of the gas phase temperature during cloud combustion. Equations used for the simulation of the AlO emission spectra are given below.

The specific intensity assuming that medium is optically thick,

$$I_\lambda(T) = B_\lambda(T)\left[1 - e^{-\alpha'_\lambda l}\right] \quad (1)$$

The absorption coefficient corrected for stimulated emission,

$$\alpha'_\lambda = \frac{E_\lambda(T)}{B_\lambda(T)} \quad (2)$$

The spontaneous emission with medium optically thin,

$$E_\lambda(T) = \left[\frac{16\pi^3 c N_u(T)\nu^4_{v'v''}}{3(2J'+1)}\right] S_{v'v''} S^{J'\Lambda'}_{J''\Lambda''} \times 10^{-11} \quad (3)$$

The number of AlO molecules in the upper state is given by

$$N_u(T) = \frac{N(2J'+1)}{Q} e^{-\frac{E_u}{kT}} \quad (4)$$

Where $B_\lambda(T)$ the Planck black body function which is calibrated with a black body furnace at 1600K, l the geometric path length, c the speed of light, $\nu_{v'v''}$ the frequency of emitted radiation, $(2J'+1)$ the spin multiplicity, $S_{v'v''}$ the band strengths, $S^{J'\Lambda'}_{J''\Lambda''}$ the line intensity factor, N the number density of molecules, Q the partition function, E_u the spontaneous emission in the upper state, k the Boltzmann constant and T the temperature.

The method to determinate the condensed phase temperature is based on the continuous part of the flame spectra. The temperature is derived of the condensed emitters in the flame. This was achieved using the polychromatic fitting of the low resolution continuum spectra in the 400-900 nm spectral range to Planck's low.

$$L_\lambda(T) = \varepsilon(\lambda,T) \times \frac{C_1}{\lambda^5} \times \frac{1}{e^{\frac{C_2}{\lambda T}} - 1} \quad (5)$$

$\varepsilon(\lambda,T)$ is the spectral emissivity of the condensed emitter, if the emitters are considered like « grey », the factor $\varepsilon(\lambda,T)$ is a constant inferior or equal to 1, so independent of wavelength. The resulting curve is function of $\frac{1}{\lambda^5}$.

Nevertheless, if the condensed phase emitters are not considered like « grey », so for fine aluminium oxide particles (d<1 µm) witch are the main source of continuum spectra, the emissivity is not equal to 1. It can be considered that for such small particles the light emission cross-section decreases with wavelength as $\frac{1}{\lambda}$, [3]. At the same time the authors claim that the absorption index of the molten bulk aluminium oxide also decreases as $\frac{1}{\lambda}$ in the 400-900 spectral range. The resulting factor $\varepsilon(\lambda,T)$ is proportional to $\frac{1}{\lambda^2}$ and hence the luminance proportional to $\frac{1}{\lambda^7}$. They found a better accordance with these hypotheses. In the present experiments the condensed phase temperature obtained with the emissivity proportional to $\frac{1}{\lambda^7}$ gives realistic results whereas with the emissivity proportional to $\frac{1}{\lambda^5}$, results are physically unrealistic.

Two different Al powders are used in the experiments: a nanopowder (250 nm) and a micropowder (6 µm) provided by SNPE Matériaux Énergétiques.

Flame propagation velocities: results and discussion

Figure 2 shows a sequence of Al particle cloud combustion. In that sequence the time between frames is 10 ms. The first picture of the sequence shows the ignition point of the cloud due to the spark. Then the combustion front propagates both downwards and upwards along the tube axis. The upwards propagation ends quickly because of the expansion of the hot products. After a stabilization time, only the downwards propagation remains and can be analysed.

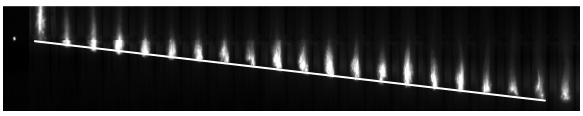

Figure 2: Sequence of Al particle cloud combustion (10 ms between frames).

Particle cloud flame propagation velocities are obtained by numerical image analysis of the combustion light emission, Figure 3a. It's presented the different phases of the analysis.

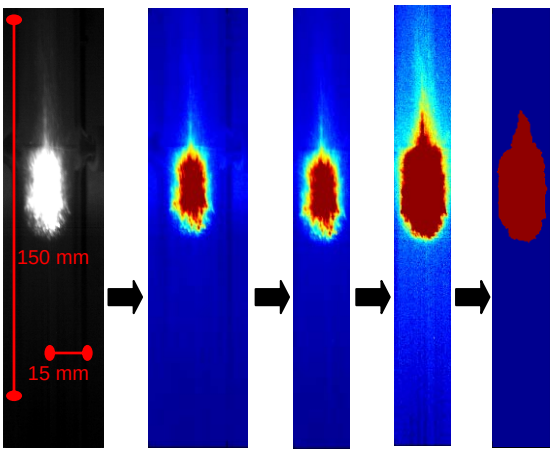

Figure 3a: Example of numerical analysis of one flame extract from the movie of the Al particle cloud combustion

Numerical analysis of each frame gives two parameters: time t and flame front position x. It's done for each frame extracted from the movie. All these data are reported in Figure 3b, which illustrates how the mean velocity is taken from the gradient of $x(t)$.

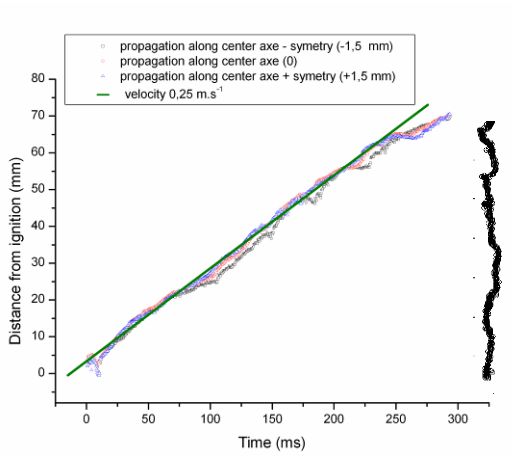

Figure 3b: Example of determination of the mean velocity for one sequence of an Al particle cloud combustion and an illustration of the path of the flame front leading point.

The experimental velocities thus obtained are collected in Figure 4.

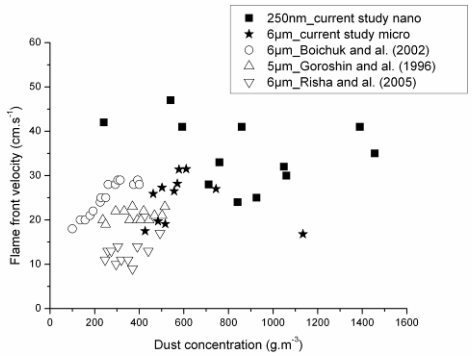

Figure 4: Flame front velocity of Al particle clouds versus dust (particle) concentration for different particle sizes, [4], [5], [6].

Figure 4 compares cloud burning velocities determined for various micro-aluminum and nano-aluminum particle concentrations in air with different data from the literature. Microparticle clouds experiments are in a good accordance with the data from the literature. The results for nanoparticle clouds give two maxima: one next to the stoichiometric concentration and the second one at a higher concentration. The second maximum can be due to the increasing total area of particle surfaces when the particle concentration rises. The experiments indicate that for the same global concentration the nanoparticle clouds burn faster than microparticle clouds.

Flame temperature: results and discussion

Figure 5 presents different stages of the analysis of emission spectra from microparticle combustion. First is the initial spectrum obtained after the combustion, then the spectrum is divided by the apparatus function and finally the continuous spectrum is subtracted to use the AlO spectrum for simulation.

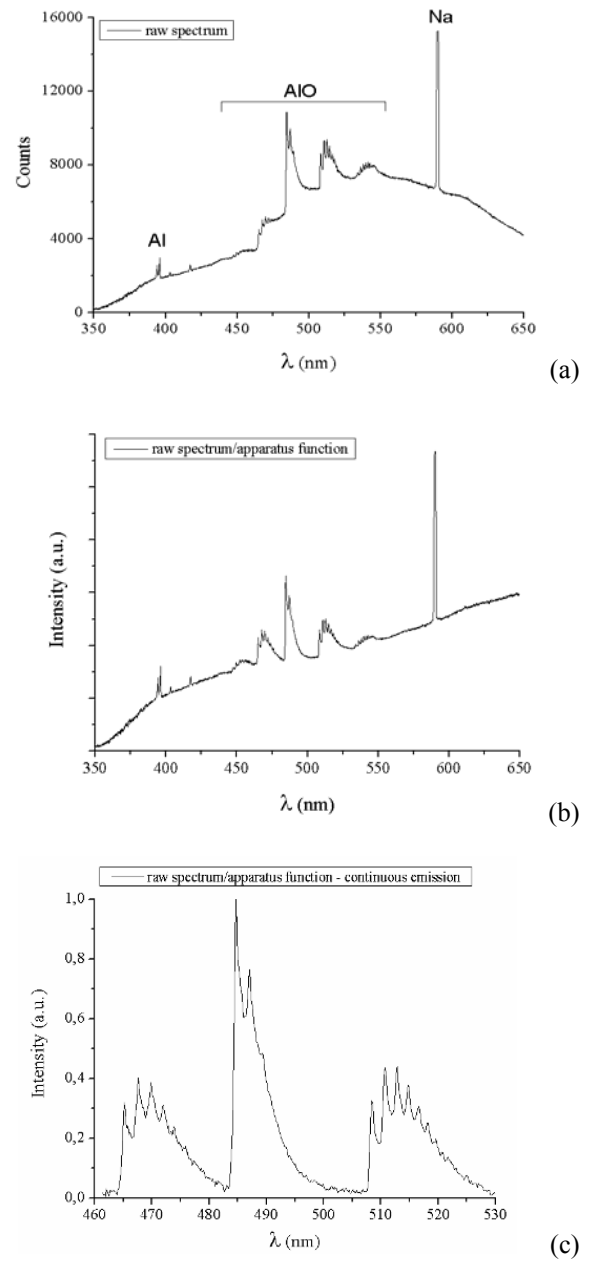

Figure 5: Different stages of the analysis of emission spectra from microparticle combustion.

The simulated AlO emission spectra are in a good accordance with the experimental spectra and give a constant value of 3300 K for the microparticle cloud combustion and 2900 K for the nanoparticle cloud combustion.

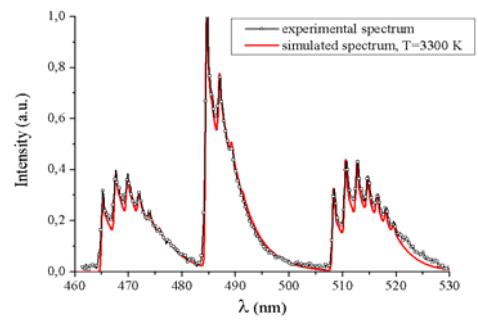

Figure 6a: Experimental and simulated AlO emission spectra during aluminium microparticle cloud combustion in air.

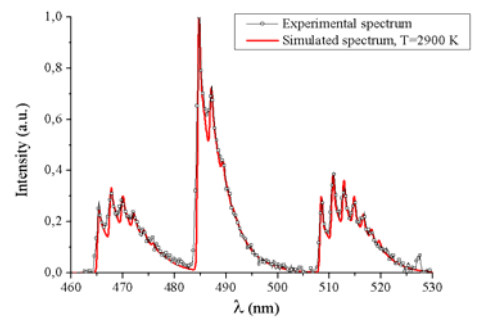

Figure 6b: Experimental and simulated AlO emission spectra during aluminium nanoparticle cloud combustion in air.

The condensed phase temperature is also exanimated in this study. Two curves are shown in Figure 7, one of micro-sized powder and the other of nano-sized powder. Each point represents the temperature of condensed emitters present in the flame for different time. The measurement zone is fixed and the flame propagates so the measurement position is along the axis of the flame. The two curves have the same profile, the temperature increases in the beginning and after a stabilisation decreases at the end. The first phase is the increasing which is fast in both cases corresponds of the ignition zone. It is clear that the distance between the unburned and the burned mixing is small. The second phase is the constant temperatures which correspond of the burning zone. Then the last phase is the decreasing of the temperature. This phase is linked with the tail of the flame. The tail of the flame is longer in the case of the nano-sized powder than in the case of micro-sized powder. The resulting temperature decreases slower with nano-sized powder than with micro-sized powder. The temperature rise faster to the maximum in the case of nanoparticle cloud combustion but that maximum is lower than in the case of microparticle cloud combustion. But the decreasing is faster for the nanoparticle cloud combustion.

The maximum temperature of the condensed phase and the temperature of the gas phase are the same for microparticle cloud combustion. Whereas for the nanoparticle cloud combustion, the temperature of the gas phase is 280 K lower than the condensed phase temperature.

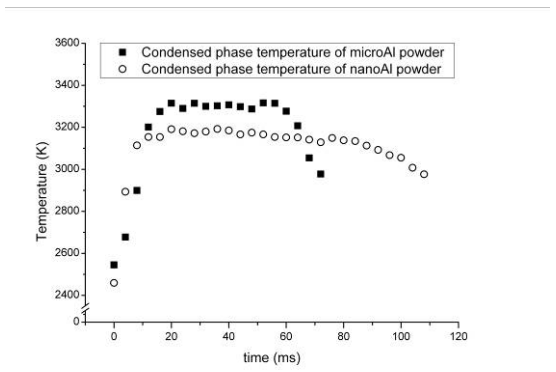

Figure 7: Condensed phase temperature of micro- and nano-particle cloud combustion in air.

Conclusions

The present results show that nano-sized Al particle clouds burn faster than micro-sized particle clouds for the same global particle mass concentration in air. The cloud flame propagation velocity depends also on the particle concentration. The global evolution of the velocity versus concentration of nano-sized and micro-sized Al particle clouds show different trends; the nano-sized Al particle clouds show two maxima whereas the micro-sized particle clouds only one. The simulated AlO emission spectra give a constant value of 3300 K and of 2900 K for the nanoparticle and for the microparticle cloud combustion, respectively. The condensed phase temperature during the microparticle cloud combustion is higher of 180 K than during the nanoparticle cloud combustion.

Acknowledgments

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