STUDIES ON THE BURNING OF COMPLEX ALUMINUM PARTICLES

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Abstract

In this work, two different techniques are used to reduce the burning times of aluminum particles with the ultimate goal to improve performance of solid propellants. One method is to coat the particles by nickel, and the second is to decrease the particle size to nanometric scale.

A thin coating of Ni on the surface of Al particles can prevent their agglomeration and at the same time facilitate their ignition, thus increasing the efficiency of aluminized propellants. In this work, ignition of single Ni-coated Al particles is investigated using an electrodynamic levitation setup and laser heating of the particles. The levitation experiments are used to measure the particle ignition delay times and burning times at different Ni contents in the particles.

Decreasing the size of Al particles increases their specific surface, and hence decreases the burning time of the same mass of particles. In this investigation, a cloud of Al nanoparticles suspended in a tube is ignited by an electric spark. The cloud experiments are used to measure the flame front propagation velocity for different particle sizes and different coatings.

The results and their analysis show that both methods reduce the Al burning time. Ni coating reduces significantly the ignition time of micro sized Al particles and hence the total burning time compared to non-coated particles. The nano-sized particle clouds also burn faster than micro-sized Al particle clouds.

Introduction

The previous studies have shown that the use of Ni-coated Al particles may improve combustion performances of aluminized propellants. For example, coating Al particles with a thin (10-100 nm) film of Ni reduces particle agglomeration [1, 2]. This effect is explained by the fact that the melting point of Ni (1728 K) is much higher than that of Al (933 K). The solid Ni layer on the surface of Al droplets prevents their coalescence and agglomeration in the surface layer of burning propellant or in the gas phase. Paradoxically, at the same time there are some indications that Ni-coated Al particles ignite easier than original Al particles. For example, studies on flame propagation in clouds of Ni-coated Al particles in air revealed an increase in the front velocity by a factor of 4 as compared with uncoated Al particle clouds [3]. The authors explained this effect by the reduction of the ignition delay time of the particles due to thermal stress cracking and peeling of solid Ni shell followed by easy ignition of the bare Al core. Note, however, that the exothermic chemical reactions between Al and Ni [4] may also promote ignition.

The combustion of single Ni-coated Al particles (36-63 µm, 51 wt.% Ni) in air, O_2, CO_2, and Ar was studied previously [5]. The particles were levitated in an electrodynamic chamber at room temperature and ignited by a CO_2 laser. A number of interesting results related to the influence of Ni coating were obtained, such as two-stage burning in air. The ignition delay time in the oxidizing atmospheres was nearly identical in all oxidizing atmospheres.

In addition, the available literature also shows that Al nanopowders can significantly improve the performance of some energetic materials, especially for propulsion applications. Solid propellants containing so-called Alex nanoscale powders exhibit burning rates much higher (in some cases as much as 5 to 20 times higher) than the propellant formulations containing regular Al powder [6]. Further, in a hybrid rocket, HTPB-based solid-fuel formulations containing Alex and WARP-1 nanopowders demonstrated mass burning rates up to 50% higher than the baseline formulations with regular Al [7]. These superior performance characteristics for energetic compositions containing Al nanoparticles instead of conventional microsize particles are based on the higher specific area and reactivity of nanoscale powders.

In the present work, combustion of levitated and laser-ignited Ni-coated Al particles is studied over a wide range of Ni mass fractions, from 0 to 14 wt.%. Note that only particles with relatively small content of Ni are of interest for propulsion applications. The emphasis is on the measurements of the ignition delay time and of the combustion time for particles with different Ni mass fractions and on comparison with the original Al particles.

The second part of investigation is related to combustion of Al nanoparticles injected into a tube and ignited by an electric spark after formation of cloud. The nanoAl particles can also be coated with different materials to prevent agglomeration. The cloud experiments are used to measure the flame front propagation velocity for different sizes and different coatings of the Al particles.

Experimental Techniques

Experiments on the combustion of single Ni-coated Al particles in air under normal conditions (room temperature, pressure 1 atm) were conducted using the electrodynamic levitation facility of LCSR-CNRS (Figure 1).

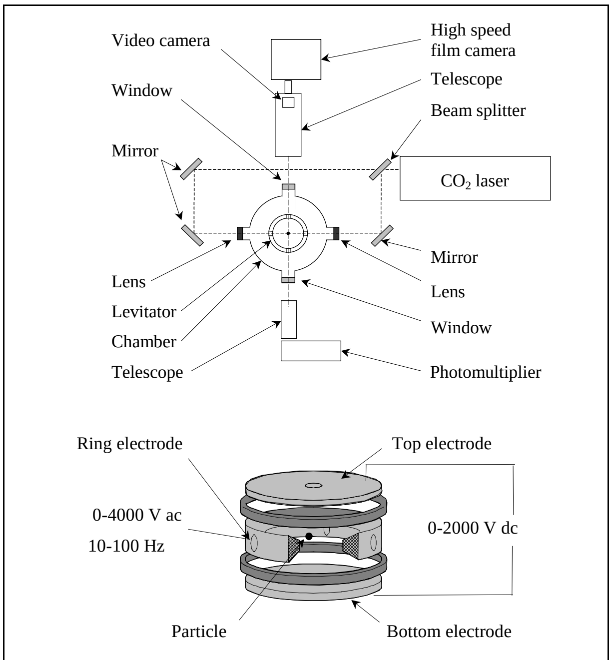

Fig. 1.Schematic diagram of the experimental setup for studies of the combustion of levitated particles; the bottom image shows the design of the levitator.

The experimental procedure and main parts of the apparatus are described earlier [8-10]. Briefly, the electrodynamic levitator allows suspending single metal particles of initial diameters in the range 15-150 µm. The levitator is placed inside a hermetic chamber equipped with a particle injection system, which permits experiments in different atmospheres and pressures. The single levitated particle is ignited by using a 50 W CO_2-laser (Synrad 48) with the beam split and focused onto opposite sides of the particle. The focusing is accomplished with two ZnSe lenses. Micrometric measurements show that the beam diameter in the centre of the levitator is ~340 µm. Prior to ignition, a video camera and a long-distance microscope (Questar QM100) are used to adjust the position of the levitated particle at the focal point of the laser. The laser is interrupted at particle ignition. A photomultiplier measures the light emission intensity of the burning particle, variation of which allows the determination of the particle ignition delay and burning times.

Figure 2 shows the experimental setup used for studying the combustion of nanoparticle clouds in air under normal conditions.

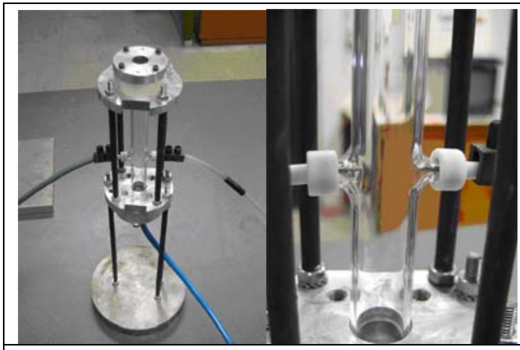

Fig. 2.Experimental set up for studies on combustion of nanoparticle clouds

The combustion chamber is essentially a quartz tube (inner diameter 14 mm, height 170 mm), the bottom end of which is closed by a porous metal plate, and the top end is closed by a paper (cellulose) film. The powder sample (13 mg corresponds to stoichiometric reaction with air inside the tube) is placed in the bottom of the tube. Suspension of particles and formation of the cloud inside the tube is achieved using an electrical valve which creates pulsed air flow in the tube to carry and disperse the particles. After suspension, the cloud is stationary due to low sedimentation velocities of Al nanoparticles, 10^{-5} – 10^{-3} cm/s. The particle cloud is ignited by a spark. The combustion front propagates upwards along the tube. Note that the use of semi-open tubes ensures combustion at constant pressure, which is attractive for measurements of laminar burning velocity.

A high-speed video camera Phantom V5.0 (1000 fps at maximum resolution of 1024 x 1024 pixels^2) is used for flame front observations and propagation velocity measurements. The laminar burning velocity of the combustion front is determined for different nanoAl samples by image analysis.

For the levitation experiments, Al particles 32-40 µm in size, from The Metal Powder Company Ltd. (Madurai, India), coated by Ni at Technion-Israel Institute of Technology are used. The six powder lots with different Ni loadings, i.e. with different thicknesses of Ni coating, were produced. Table 1 (columns 1-6) summarizes the data on the mass fraction of Ni in the powder lots determined with atomic absorption spectroscopy and on the coating thickness calculated for the mean diameter with the assumption that the particles are ideal spheres with uniform Ni coating. In addition, the coating thickness for the particles with 4.33 wt % Ni was measured using Auger electron spectroscopy (AES) in Technion. The value of 56 nm, obtained with the standard assumption that the Ni layer depth corresponds to half of the maximum Ni concentration, is in reasonable agreement with the estimates. Further, AES analysis of the initial Al particles clearly shows that the thickness of the oxide film on their surface is about 10 nm.

Ni, wt. %	0.93	1.87	3.11	4.33	9.10	14.4
Thickness, nm	17	35	58	81	180	301
Particle size, µm	32-40	32-40	32-40	32-40	32-40	32-40

Table 1. Ni mass fractions and coating thicknesses in the used Ni-coated Al particles.

For cloud burning experiments, three different powders were used: Alex-type Al nanopowder produced at the Institute of High Current Electronics (Tomsk, Russia), Al nanopowder produced by Gen-Miller method and coated by 2 - trimethylsilyl at the Institute for Energy Problems of Chemical Physics in Moscow (the mean particle size for both nanopowders is 150 nm), and a micro-scale Al nanopowder (6 µm) obtained from SME.

Experimental Results

Figure 3 shows typical profiles of the light emission intensity measured during ignition and combustion of Al (3a) and Al/Ni (3b) particles. The laser is interrupted when the light emission intensity reaches a threshold, which is fixed in all experiments. The time from starting the laser to its interruption is determined as the ignition delay time. The time from this interruption of laser to the beginning of stabilization of the light emission intensity is the combustion time.

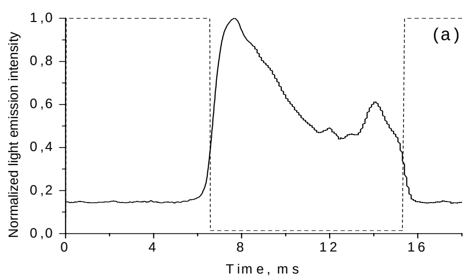

Fig. 3a. Light emission during ignition and combustion of Al particles in air. The dashed line shows the laser monitoring sequence (0 = off, 1 = on).

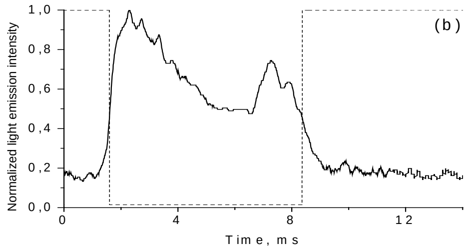

Fig. 3b. Light mission during ignition and combustion of Al/Ni (14.4 wt.% Ni) particles in air. The dashed line shows the laser monitoring sequence

Figure 4 shows the measured ignition delay time, the combustion time and the total combustion time of particles with different contents of Ni. Each data point was obtained by statistical analysis of 20-40 experiments. It is observed that the ignition delay time significantly decreases with increasing the Ni mass fraction from 0 to 3% and changes only slightly with further increase in the Ni content. The data for low coatings are characterized by large scattering. This implies that these powders contain particles with partially coated surface (Figure 5). Indeed, it is difficult to reach high coating quality for low content of the coating material. However, the obtained data allow clear identification of two characteristic values of the ignition delay time: one for the original Al particles, and the second, much shorter, for Al particles with Ni coating at 3-14 wt.% Ni. The data of the combustion time show no clear indication of variation, and the total burning time decreases due to the decrease of the ignition time.

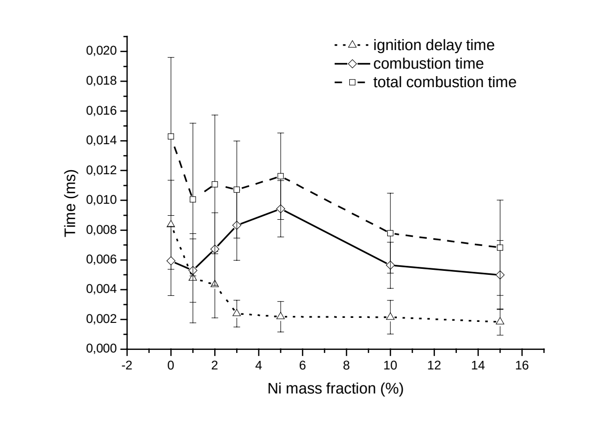

Fig. 4. Ignition delay, combustion and total combustion times of Ni-coated Al particles in air as a function of Ni mass fraction.

Figure 5 represents two SEM pictures of the coating by Ni of aluminum particles.

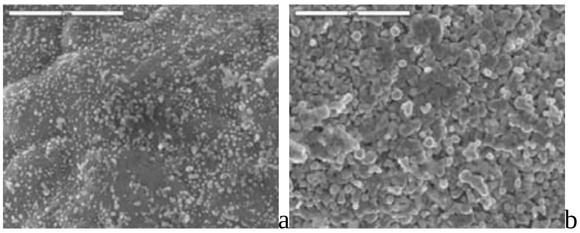

Fig. 5. SEM visualization (a) 2 wt % Ni coating (b) 10 wt % Ni coating on Al particle.

Figure 6 shows two successive images of particles cloud combustion obtained with the high speed video camera and the same images after image analysis. The difference between two flame's front positions divided by the time between two images (322 µs for Figure 6) gives an estimate of flame propagation velocity. The final flame velocity is calculated using the successive images corrected by the expansion factor.

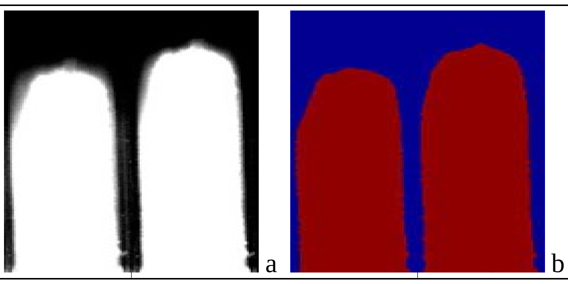

Fig. 6. (a) Two successive images of the cloud flame (b) Digitized images of the same sequence

Figure 7 shows the measured cloud flame front velocities for different initial conditions (7a, 7b) and for different samples (7c, 7d).

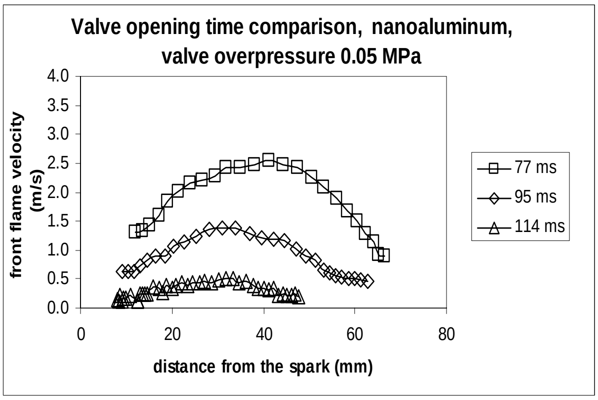

Fig. 7a. Flame front velocity of nanoAl particles cloud for different valve opening time.

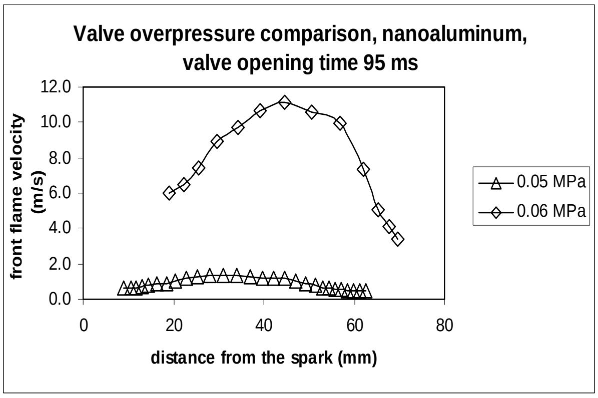

Fig. 7b. Flame front velocity of nanoAl particles cloud for different valve overpressure.

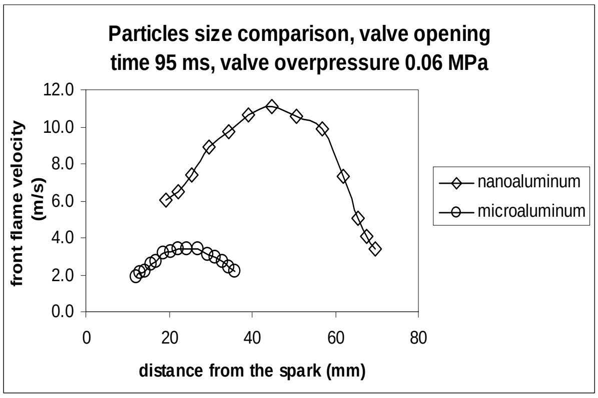

Fig. 7c. Flame front velocity of nanoAl and microAl particles cloud

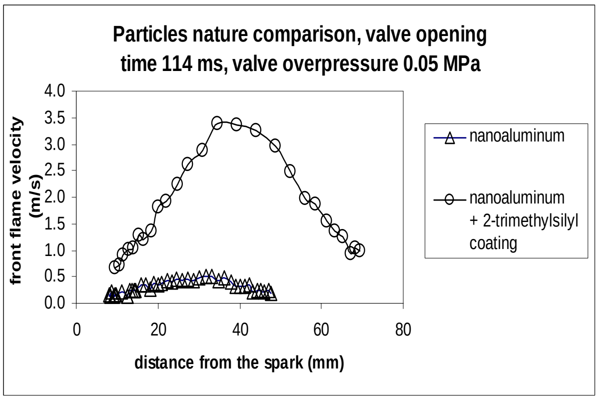

Fig. 7d. Flame front velocity of coated and no-coated nanoAl particles cloud.

Discussion and conclusions

Figure 4 represents the ignition delay, combustion and total combustion times of Ni-coated Al particles in air as a function of Ni mass fraction. The prior study [11] shows that the ignition delay time of single Ni-coated Al particles in air is significantly shorter than for original Al particles, obviously due to the lower ignition temperature of coated particles. The total combustion time also decreases. There are, however, some variations for the burning time after ignition depending on the coating thickness which should be explained further referring to the quality of coating for low coating mass and to the role of nickel burning.

Figure 7 represents the cloud flame front velocity for various samples in different initial conditions of suspension. The velocity curves present a parabolic shape versus distance from ignition. The several factors can be enumerated to explain this behavior. At the beginning the cloud can have a residual velocity due to the pulse which permits the suspension of the particles. Second, this behavior can be caused by inhomogeneous particle distribution in the tube. The particle concentration can be too high or too low at the beginning near the ends of the tube but in an optimum proportion in the middle of it.

The comparisons show different behavior in various conditions. Figure 7a and Figure 7b demonstrate that the cloud burning velocity increases with increasing the valve overpressure and with decreasing the valve opening time. One possible explanation is the formation of agglomerates during the suspension process. The agglomeration is indeed favored by a long time of contact between the particles. The agglomerated particles spend more time to melt and to vaporize; therefore the velocity of the cloud flame is lower. Figure 7c and Figure 7d illustrate the effect of coating. Both samples have the same size distribution but one of them is composed of Al nanoparticles coated by 2 - trimethylsilyl while the other is composed of non-coated Al nanoparticles. The coating prevents agglomeration of the nanoparticles in the cloud, so the particles burn easier. The flame front velocity for the coated sample is higher by a factor of 7. The particle size changes the cloud burning velocity too; the burning velocity of the cloud flame of nano particles is by 3 times higher than for the microsize particles.

Additional experiments are ongoing to characterize the ignition of coated micron sized Al particles and the burning regimes of coated nano Al particles in better manner.

Acknowledgements

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