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Combustion times and emission profiles of micron-sized aluminum particles burning in different environments

Robert J. Gill, Carlo Badiola, Edward L. Dreizin *

Otto H. York Department of Chemical, Biological, and Pharmaceutical Engineering, New Jersey Institute of Technology, Newark, NJ 07102, United States

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ABSTRACT

Optical signatures and combustion times for micron-sized aluminum particles are measured in oxidizing environments including nitrogen mixtures with oxygen, carbon dioxide, and water. Particles in a room temperature oxidizing gas stream are fed into a CO_2 laser beam where they are ignited. Prior to entering the CO_2 laser beam, each particle crosses a second, low energy laser beam and produces a scattered light signal used to determine the particle size in real time. The correlation between the measured particle sizes and their burn times produces an experimental trend that is compared to various correlations reported in the literature. In addition to the burn time measurements, detailed optical signatures are recorded for micron-sized aluminum particles burning in different environments. For aluminum burning in water vapor, the optical signature of the particle is substantially weaker than in other environments, possibly indicating a primarily surface oxidation. It is shown that semi-empirical and widely used τ_b–D^n expressions for the particle burn time, τ_b, as a function of its diameter, D, are inaccurate for the conditions that are different from those used to establish respective trends initially. It is observed that the effect of oxygen concentration on combustion time of micron-sized aluminum particles is weak when oxygen concentrations exceed 21%. Combustion times increase substantially for lower oxygen concentrations. Aluminum particle combustion times are substantially longer than predicted for experiments with water and carbon dioxide oxidizers. For all environments, the observed effect of particle size is relatively weak and the exponents in the descriptive D^n relations appropriate for the current experiments vary approximately from 0.3 to 1. The exponent close to 0.3 adequately describes current results for those oxidizing environments for which heterogeneous reactions appear to dominate.

1. Introduction

Aluminum is an important energetic component of many solid propellants, explosives, and pyrotechnic formulations [1–4]. One critical parameter of aluminum combustion, universally important for all applications, is the particle burn time, τ_b, as a function of the particle diameter, D. For practical purposes the burn time is commonly expressed as a power law $\tau_b \sim D^n$, with the exponent n and pre-exponent factor depending on the oxidizing environment, temperature, and pressure, e.g., [5–8]. Various D^n type trends were reported by different authors based on a diverse set of experiments. Generally, laboratory experiments in well-characterized environments reported in the literature can be broadly divided into two groups: experiments using individual metal particles, e.g., [9–14], and experiments employing aerosolized powders or clouds, e.g., [7,15–19]. For individual particles, the measurements of combustion times are often direct, while indirect methods and data analyses are used to extract the information on particle burn times or burn rates from the cloud combustion experiments. It is also worth noting that single metal particle combustion experiments in general, and direct measurements of combustion times for individual aluminum droplets in particular, were restricted to relatively large particles, with sizes 50 μm or greater. However, most practical applications deal with finer aluminum powders with particle sizes on the order of, or finer than, 20 μm. It is also interesting that in many experimental configurations, aluminum particles burn in combustion products of hydrocarbon fuels. In such cases, the oxidizers are mixtures of CO_2, H_2O, and O_2 in various proportions. While this situation imitates some practical applications, the specific oxidizer mixtures produced in laboratory burners and in practical energetic formulations differ from one another substantially. Laboratory experiments with mixed oxidizers are also not particularly useful for extracting the information about efficiency of individual oxidizing species, which is required to model the practical configurations. The information on aluminum combustion in CO_2 and H_2O is very limited [9,20,21], with most

* Corresponding author. Fax: +1 973 642 4282.
E-mail address: dreizin@njit.edu (E.L. Dreizin).

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data coming from experiments in mixed oxidizers, where the effects of different oxidizers are somewhat difficult to uncouple.

Considering the limitations of the available data on aluminum combustion, the goal of the current work was to directly measure optical signatures and burn times of individual, micron-sized aluminum particles. The focus of these experiments was to establish a direct correlation between particle diameters and their optical signatures and burn times for particles under 20 μm. The measurements were performed in well-characterized oxidizing environments with individual oxidizers including O_2, CO_2, and H_2O. These measurements are expected to serve as a foundation for development of a mechanistic aluminum combustion model. The model should account for multiple processes occurring in aluminum combustion and is not expected to be limited to a quasi-steady description. At the same time, the model should be relatively simple to enable its implementation in practical calculations. The measurements reported in this paper are expected to help to identify the most important reaction mechanisms and processes to be included into the mechanistic model of aluminum combustion.

2. Experimental

2.1. Apparatus

The experimental setup used in this study is schematically shown in Fig. 1. The apparatus, experimental procedure, and data processing steps have been described in detail in a recent publication [22], and only a brief summary is presented here for completeness. Metal powder, comprised of spherical particles, was fed by an electrostatic particle generator [7,19,22] so that a narrow, vertically rising particle jet with a low number density is produced. The particles were carried by an oxidizing gas stream. The particle jet crosses two laser beams. First, particles intercept a 785-nm-laser beam. For each particle, the scattered light intensity is proportional to its area and is used to measure the particle diameter. Two millimeters above the 785 nm laser beam, the particles traverse a focused CO_2 laser beam (~0.3 mm beam waist) in approximately 0.5 ms (i.e., particle velocities ~0.6 m/s). In the CO_2 beam, particles are rapidly heated and ignited. Emission from the incandescent and burning particles was measured to determine the particle burn times. The emission was collected by a wide angle quartz fiber, passed through a 500 nm band-pass filter, detected by a photomultiplier tube, converted to a digital signal, and stored for further analysis.

There are three significant modifications to the apparatus previously described in Ref. [22].

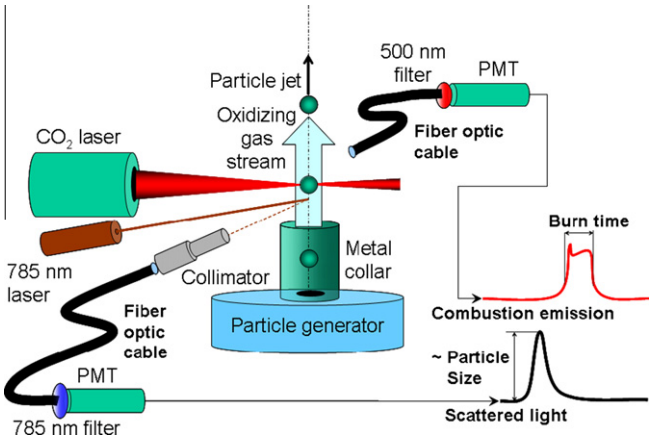

Fig. 1. Schematic diagram of the experimental setup.

First, a set of small inner diameter telescoping tubes, labeled "collimator" in Fig. 1 were attached to the fiber optic viewing the 785 nm scattered light. The field of view of the detection system was reduced to <4.5° (full angle) and these tubes eliminated all emission produced by incandescent or burning particles from reaching the scattered light detector.

Second, experiments were performed in various oxidizing environments. The compositions of the oxidizers included mixtures of N_2/O_2, N_2/CO_2, and N_2/H_2O (all at 1 atm). A thin-walled aluminum collar ("metal collar" in Fig. 1) was added to the particle generator. This restricted outside air from mixing with the oxidizing gas streams. This collar is 8 mm in height and it has an outer diameter of 8.9 mm (inner diameter = 6.4 mm). High purity, dry gases were used for these experiments. Gases were metered through needle valves and flows were measured with mass flow meters (Alicat Scientific; Model M200SCCM-D) to ±1% accuracy. Carbon dioxide and water vapors interfere with the operation of the particle generator; so for experiments with these oxidizers, particles were fed with a pure nitrogen stream, and oxidizers were supplied in a shroud jet [22,23] not shown in detail in Fig. 1. Detailed calculations using FLUENT software, for the gas flows in this apparatus, show that the inner and outer gas streams are well-mixed at the height where the particles intercept the CO_2 laser beam. These results are presented in Ref. [23] where the same experimental configuration was employed to study ignition of aluminum particles in different oxidizers. For oxygen/nitrogen gas mixtures, both the inner gas flow passing through the particle generator and the shroud jet had the same gas composition. Individually metered oxygen and nitrogen gas streams were mixed by passing them through a 3.0 m length of plastic tubing (1/4 in. od). To generate water vapor, a calibrated peristaltic pump (Control Company; Model 3384-CC) fed liquid water (3.3 mg/s) into a tube furnace held at a temperature of 200–250 °C. The gas transfer lines and a top flange of the particle generator were maintained at 120 °C to prevent condensation.

Specific environments used in experiments, gas flows, and calculated flow velocities are shown in Table 1. Particle flow velocities along the centerline were measured using square-wave modulated (500 Hz; 50% duty cycle) green laser sheet illumination. This yielded particle velocities of about 0.6 m/s at 5 mm above the collar rim (13 mm above the particle generator outlet) and these velocities agree with those estimated from the offset time found when correlating the scattering peaks with the corresponding ignition peaks (see below for details).

Lastly, a 14-bit data acquisition board (DAC, National Instruments, Model PCI-6133) replaced the 12-bit, sequential DAC used in prior work. The new DAC has eight independent D/A converters and a maximum sampling rate of 3.0 MS/s (0.33 μs per sampling event). In the current experiments, the sampling rate was 100 kS/s; or 10 μs per data point; except for the experiments with air, where the sampling rate was increased to 500 kS/s. For each scan, the time during which sequences of pulses of both scattered light and light produced by incandescent particles were continuously collected, was 10 s. Typically, 50–150 scans were collected for a set of experiments with a selected oxidizing gas mixture.

2.2. Materials

Spherically shaped micron-sized aluminum particles were used for these experiments (Alfa Aesar, 10–14 μm nominal particle size). Prior to experiments, the powder was dried in a glass vial at 65–85 °C under vacuum for more than 2 h. The vial was quickly capped after opening the vacuum oven and after a cooling period, the sample vial was roll-milled (i.e., milled with an aluminum or Teflon solid rod, 1 cm diam.) for approximately 20 min. This procedure reduced the number of agglomerated particles, but did not