Ignition characteristics of laser-ablated aluminum at shock pressures up to 2 GPa

Kyung-Cheol Lee, Tsubasa Taira, Goon Mo Koo, Jae Young Lee, and Jack J. Yoh

1Department of Mechanical and Aerospace Engineering, Seoul National University, 1 Gwanakro, Gwanakgu, Seoul 151-742, South Korea
2Division of Applied Physics, Hokkaido University, Sapporo, Japan
3Department of Aerospace Engineering, University of Maryland, College Park, Maryland 20742, USA

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The ignition of aluminum particles under high pressure and temperature conditions is considered. The laser ablation method is used to generate oxide-free aluminum particles exposed to pressures ranging between 0.35 and 2.2 GPa. A continuous wave CO2 laser radiation heats the surface of the aluminum target until ignition is observed. We confirm ignition by a spectroscopic analysis of AlO vibronic band of 484 nm wavelength, and the radiant temperature is measured with respect to various pressures for estimating the heating energy for ignition. The ignition characteristics of the oxide-free aluminum particles exposed to extremely high pressures are reported. © 2014 AIP Publishing LLC.

I. INTRODUCTION

Aluminum (Al) powders are often added during high explosive (HE) synthesis to modify the responsive pressure impulse and temperature profile of otherwise well-characterized detonation behavior of a HE. The time scale of energy release for reaction of Al particles is typically several orders of magnitude slower than that for the HE. Thus, understanding the reactivity of aluminum particles exposed to a hot product gas environment from an initial detonation of high explosive is critical for making full use of such energy efficient metal additives.

The combustion of aluminum particles has been studied extensively for decades, mostly at pressure conditions below 10 MPa as these experiments were conducted in the pressure chamber and shock tube where there is known relation between the aluminum particles and the ambient pressure. On a contrary note, a metal combustion in the detonation of aluminized high explosives and propellants involve local pressures reaching several giga pascals (GPa). Often times, it is difficult to assure ideally detonative conditions or smooth detonation transition from an ignition stimulus by the presence of the aluminum additives.

Two distinct burning regimes, namely, kinetics-limited and diffusion-limited regimes based on the particle size, for aluminum particles have been identified from the past studies performed at atmospheric pressure. Particles larger than ~20 μm combust in a diffusion-limited manner1 with a detached diffusion flame close to the particle surface.2-4 The reaction kinetics is fast and the overall burn time is limited by the diffusion of oxidizer. The dissociation temperature (~3300 K) of the oxidized particle controls the temperature peaks within the flame region.2 As the particle size is reduced, diffusion becomes faster and the flame moves closer to the surface, heterogeneously.5 A peak temperature in the flame exceeds the aluminum boiling temperature (~2800 K). However, combustion of a particle less than 10 μm in size becomes strongly kinetics-limited. The oxidizer diffuses quickly into the particle core through the shell, with the reaction kinetics being the limiting factor. The peak temperature is achieved inside the particle in this case, but combustion temperature remains below the boiling point.6

Although it has been known for some time that the pressure will play a little role in particle ignition and combustion, only a limited number of experimental studies have examined this important role. In a recent effort by Marion et al.,2 they measured the pressure dependence of heating and burning time of 40 -μm aluminum particles in air in the pressure range of 0.1–4 MPa using a laser for ignition. The high speed images are used for estimation of heating and burning time. The heating time for ignition and ignition delay are independent of pressure and are constant. The burning time increased weakly for increasing pressure. Nevertheless, experimental pressure range as considered in their work is much lower than the explosion pressure of aluminized explosive in a GPa range.

The combustion of aluminum particles has been studied using numerous laboratory-scale methods that considered particle size, oxidizer concentration, pressure, relative velocity, and temperature.1,5,7-9 Using a gas burner, aluminum particles can burn in the exhaust gas products of the flame, consisting of H2O and CO2 as the oxidizing species.4,10-12 This creates an atmosphere similar to that of a burning solid propellant. Flame propagation in a mixture of aluminum dust and gaseous oxidizer was also examined.13-18 Tanguay et al. studied burning of 10–100 μm particles in a high-speed detonation flow of a stoichiometric hydrogen/oxygen mixture at atmospheric pressure.19 At relative velocities of 100–200 m/s, burning times were found to increase as d−0.5, which is consistent with a kinetics-limited combustion, rather than the classical d2-law or Beckstead’s d1.8-trend.20 A polychromatic fitting of the spectrally dispersed emission was used to estimate the combustion temperature in the range of 2200–2800 K.

Laser ignition is another method that offers readily adjustable energy transfer to achieve a heating rate of interest (~107 K/s for nanosecond lasers) and to probe the particle

Email: jjyoh@snu.ac.kr. Tel.: 82-2-880-9334.
combustion. Bucher et al.\textsuperscript{3} used a 150 W laser to ignite 230 \( \mu \)m Al particles in pure N\textsubscript{2}O, pure CO\textsubscript{2}, and in a mixture of O\textsubscript{2}, N\textsubscript{2}, Ar, and He. Dreizin et al.\textsuperscript{21,22} studied the heating and ignition of micron-sized (3–8 \( \mu \)m) aerosolized aluminum particles at varied heating rates in the range of 10\textsuperscript{6} K/s using a CO\textsubscript{2} laser. Lasers were also used to probe the size effect in the initiation of pressed powders of high explosives\textsuperscript{23–26} and the correlation between size effect, light scattering, and critical diameter of detonation. Laser ignition of pyrotechnic composites was examined by Refs. 27–29. The effects of stoichiometry, particle size (nano vs. micro), and laser energy density on the ignition delay and burning time were studied. The influence of surface optical properties on the size effect was also examined.

In this paper, our aim is to investigate the ignition characteristics of aluminum particles exposed to the shock pressure (0.3–2.2 GPa) and radiant temperature (5000–9300 K) conditions. A laser-induced ablation technique is adopted for generation and dispersion of oxide-free aluminum particles in an air with the pressure approaching those in the early expansion of a detonating explosive. First, the generation and dispersion events of particles are visualized to analyze the behavior of particle cloud, and then the ablation is analyzed to predict the resulting pressure range. Second, the life time of the plasma induced by a pulsed laser radiation only is compared with that produced by both pulsed laser and continuous wave CO\textsubscript{2} laser. The cloud is ignited by a CO\textsubscript{2} laser, and the emission spectroscopy is used to confirm the ignition and combustion of aluminum cloud. The CO\textsubscript{2} laser energy provides a heat source or radiant heat for the ignition. The present laser-based ignition approach offers a novel control over the high pressure condition, and in particular, the CO\textsubscript{2} beam provides a desired thermal condition suitable for a subsequent aluminum ignition. A high-pressure equation of state and relevant chemical kinetics for aluminum particles are required for pursuing a corresponding numerical simulation which would accompany the present experimental study in a separate paper.

II. EXPERIMENTAL METHODS

A. Generation of oxide-free aluminum particles at severe pressure environment

Laser ablation technique has been proven effective for generating micro/nano-size particles\textsuperscript{30–32} at high pressure environment.\textsuperscript{33} Particles generated from laser ablation have a narrow particle size distribution with variable primary particle size and shape.\textsuperscript{31,32} The flexibility of the present laser ablation set up allows for a subsequent particle generation in air as well as in liquid.

A procedure for particle generation via the laser ablation is as follows. Laser is irradiated on an aluminum surface, and a small amount of aluminum is ablated due to the phase explosion of a targeted surface. Plasma is formed and then starts to cool down. The molten particles are ejected from a resulting crater. Figure 1 is a visualization scheme of the ejected particles using the scattering method and the imaging of the aluminum particles ejection.

Aluminum is ablated by 1064 nm Nd:YAG pulsed laser as shown in Fig. 1(a). The spot diameter is 1 mm and the pulse duration is 9 ns. The particles are ejected from a molten crater, and a 532 nm pulsed laser with a time delay is irradiated on the ejected particles for visualization (Fig. 1(b)). Light is scattered by the lifted particle cloud and is collected by a camera.\textsuperscript{34} The particle ejection image is captured at 25 \( \mu \)s after the incident 100 mJ irradiation. A target is an aluminum plate of 5 \( \times \) 5 \( \times \) 0.5 mm in size. The particle dispersion is vertical to an irradiated surface in the image.

Figure 2 shows ejection of aluminum particles at different laser energies. The bright spot on the target surface is the plasma where a high pressure is generated in the region during the ablation followed by the molten particle ejection at \( \sim \)10 \( \mu \)s after the beam radiation. The laser supported shock...
wave (LSC) is produced, followed by the aluminum particle dispersion.\textsuperscript{33} The lifetime of LSC is less than 1–2 ms, and the shock strength depends on the expansion velocity which decays slowly.\textsuperscript{35,36} In our experiment, ignition occurs within 10 µs and thus LSC pressure has an influence on the particle ignition. The strength of LSC is estimated from the laser irradiation \(I\) such that

\[
P = 1.18 \left( \frac{A}{Z} \right)^{1/3} \left( \frac{I}{10^{14} \text{W/cm}^2} \right)^{2/3} \times 10^3 \text{ [GPa]},
\]

where \(A\) is the atomic number 26, and \(Z\) is the average degree of ionization 220. Figure 3 shows resulting pressure as a function of the laser irradiance for corresponding range of laser energy between 0.1 and 1 joule per pulse. The irradiance ranges from 3 to 32 GW/cm\(^2\) with a spot size of 1 mm in diameter, resulting in the maximum pressure of 2.2 GPa.

The generated aluminum cloud and the ablation pressure establish an environment similar to that of particles exposed to a detonation condition as intended in the present investigation.

B. Ignition at high pressure

The laser ablation using a pulsed laser establishes a shock pressure condition for aluminum ignition, while a continuous CO\(_2\) laser controls the desired temperature condition. Figure 4 is a schematic of the laser and detection system for studying the aluminum particle ignition at high pressure and temperature conditions. A CO\(_2\) laser beam with 30–330 W is irradiated on a 1 mm cross sectional area of an aluminum wire. The beam is aligned with the direction of particle ejection as such to maintain the elevated temperature of the surface. The temperature is calculated from the radiant power as

\[
q = \sigma T_r^4,
\]

where \(q\) is the laser irradiance, \(\sigma\) is the Stefan-Boltzmann constant (5.67 \times 10^{-8} \text{ W/m}^2 \text{ K}^4) and \(T_r\) is the radiant temperature by the laser. Figure 5 depicts the calculated radiant temperature as a function of the laser power. For the given laser irradiance, resulting range of temperature is between 5000 and 9000 K.

The plasma induced by the ablation via the pulsed laser, aluminum melting induced by a CO\(_2\) laser, and the burning aluminum particles by a combination of pulsed and CO\(_2\) laser irradiation are all visualized by using a high speed camera and compared of their emission spectrum. The plasma from the pulsed irradiation is probed for its life time and the elements of the discharged aluminum cloud. The ignition of aluminum particles is confirmed through the tracing of the AlO band. The spectroscopic analysis herein provides an
effective means of detecting aluminum ignition and the subsequent burning.\textsuperscript{18,22,37,38,46}

We used Andor Mechelle 5000 for spectroscopic analysis. The plasma is generated by a pulsed laser beam (Continuum, Inc., Powerlite), and the plasma light is collected by a quartz lens with a 100-mm focal length, which is perpendicular to the laser direction. The plasma light collected is sent to an echelle spectrometer with 0.1 nm resolution and an ICCD (Andor iStar 1024 × 1024) to record the signal. Various time delays and 1 \(\mu\)s gate width are used for the measurements. For the detector used in the experiment, the detectable range was between 200 nm and 975 nm. The wavelength calibration using a mercury lamp was performed before the experiment. The measured peaks of mercury lamp are checked and calibrated through a comparison between the measured and the referenced wavelength.

One of the intermediate products of aluminum combustion in air is AlO. As it is produced, the specific emission band with peaks at 447, 465, 484, 508, and 534 nm appeared as shown in Fig. 6. The band heads not clearly shown are due to a continuum noise commonly associated with the CO\(_2\) beam interaction during AlO detection. However, the range of measured band is within a broad distribution of 440–560 nm wavelength including several well-known AlO bands. In particular the measured signal intensity of 484 nm band is strong enough, and the delay time of signal was 100 \(\mu\)s to suppress any unwanted plasma effect. Thus, the temporal intensity profile at 484 nm can be chosen to quantify the burning time. Also, there are few strong peaks near 400 nm and 589 nm, which are indicative of aluminum lines (394.4 nm and 396.15 nm) and the sodium lines (589.0 nm - Na D2 and 589.6 nm - Na D1). The recorded spectra were not calibrated for wavelength-dependent sensitivity of our optical system because our main interest is determination of burning time rather than determining, for example, temperature from the recorded spectra. However, Fig. 6 indicates diminished sensitivity at both the high and low end regions of our measured spectra, yet the \(\Delta v = 0\) bands allowed us to evaluate burning time.\textsuperscript{46}

In the present experiment, high pressure and high temperature environment is established by using a pulsed laser and a continuous wave CO\(_2\) laser, respectively. The CO\(_2\) laser energy required for ignition is used to estimate the radiant temperature and consequently the ignition temperature of the aluminum at high pressure condition. Confirmation of ignition of aluminum particle and measurement of the burning time are made via the spectroscopic analysis.

\section*{III. RESULTS}

\subsection*{A. Detection of ignition}

The diameter of aluminum wire used as a beamed target is chosen in such a way to allow surface temperature to rise quickly upon laser heating. The length is however kept sufficiently long to provide continuous ablative material for sustaining an ignition. Figure 7 shows ignited aluminum...
particles that depend on the initial temperature of aluminum wire. Here, the surface temperature of a target is measured by an infrared pyrometer. The exposure time is 3 s, and a total of 30 laser pulses was considered. The captured ignition pattern consists of “dots” of burning particles for wire temperature below 830 K. At above 830 K, “streaks” of burning particles are observed. The observed short burning time for “dots” is mainly due to a small mass associated with the nano-sized particles of aluminum that burn in the solid phase. Whereas, most of the micro-sized particles are produced from ablation of the heated aluminum, and thus more particles are involved in the burning. In particular, vigorous ignition is observed when aluminum wire is heated above 900 K. The particle size is sought to be responsible for such observations.

The ablation particles are then collected for their size analysis. A glass substrate is installed at 3 cm from the target even though not all dispersed particles are expected to be recovered by the substrate of 5 × 2.5 cm in size. The particles are collected from a single pulse incident when the aluminum target is heated between 800 and 900 K. Figure 8 shows size vs. number count of the particles collected from a total of 10 pulses. Collected particle samples are imaged using a SEM for particle size and number analysis.

The particle size from laser ablation is usually less than 100 nm for most metals at room temperature. In Fig. 8, majority of produced particles are within a range of 60–90 nm. Noticeably, particles of 3–4 μm in size are also produced from laser ablation of a “heated” sample as opposed to a room temperature aluminum. The ignition of these micro-particles may explain the observed ignition pattern of Fig. 7, showing a longer burning time for micro-particles of the present experiment.

The visualization of the ignition helps to confirm appearance of aluminum particle flame and to measure and compare the burning time and the ignition delay. The irradiation from 260 mJ pulsed laser alone (Fig. 9(a)) shows periods of induction and decay of the plasma. The plasma is relatively small and its strength diminished rapidly by the plasma cooling. The plasma by a pulsed laser disappeared after 150 μs, 330 W CO2 laser alone (Fig. 9(b)) shows only a small light spot and no burning at all while CO2 laser is on. Subsequently, this minute trace of emission is believed to come from the melting of a target. For combined system at 260 mJ pulsed irradiation with 330 W CO2 beam heating, Fig. 9(c) showed both plasma and burning. The initial plasma resembles case (a) of pulsed laser alone. However, plasma and burning are sustained for the duration exceeding 150 μs. The mass of particles of laser ablation starts to eject at 15 μs, and it continues beyond 150 μs. We trace 484 nm AlO vibronic wavelength using spectroscopy to confirm the ignition and burning of aluminum particles. The relative intensity ratio is obtained by dividing the measured intensity by the averaged background signal. The relative intensity ratio of AlO band (484 nm) in Fig. 9 rapidly decays beyond 10 μs when a pulsed laser alone is used. However, the signal reaches its maximum at 11 μs and is continued when CO2 beam is combined with a pulsed irradiation in case (c). If the emission spectrum of case (c) was of the plasma origin, it would have decayed after 11 μs as in case (a). But because of emission growth due to the aluminum burning, the signal strength is unchanged. Furthermore, the AIO intensity ratio spectrum for each test confirmed that the emissions do come from the aluminum burning as additionally verified through the captured high speed images of Fig. 9.

Figure 10 shows extended time images of ignited aluminum particles and subsequent burning in air during pulsed irradiation combined with the CO2 laser heating. Unlike the plasma which decays within 150 μs when pulsed laser alone is applied, burning continues well beyond 300–400 μs in Fig. 10 with particle ejection occurring at 10 μs. The concentration of particles decreases due to ejection from the burning cloud. The cloud becomes smaller with decreasing particle concentration. The burning time of aluminum particles cloud in this case is twice longer than the pulsed laser alone. One possible cause for such burn time enhancement for aluminum may come from the interaction of plasma and CO2 beam. The lifetime of plasma is extended by the reheating via the CO2 beam.

B. Ignition temperature and burning time

The equation of state (EOS) for aluminum reflects that the melting point of aluminum increases with pressure as the melting temperature of aluminum is 960 K at atmospheric pressure, and increases to 1100 K at 2 GPa. This increase of melting temperature leads to a delayed onset of evaporation, thus requiring a higher temperature or energy for inducing ignition of aluminum in air.

Pressure can be estimated for varying pulsed laser energy using Eq. (1), while radiant temperature of a CO2 laser is given by Eq. (2). A CO2 laser power reflects a desired radiant temperature for ignition in each calculated pressure condition, and then high speed camera images.
differentiate an ignition from a failure for the tests performed. Tests are repeated 10 times for constructing the error bar for each pressure condition. Figure 11 shows the resulting ignition boundary, plotted on a pressure vs. radiant temperature graph.

Accordingly, more radiant energy is needed for aluminum to ignite in higher pressure conditions. This means that the ignition temperature also must increase at a higher pressure, and a faster heating rate is necessary for the ignition of aluminum particles. Thus, one can expect a sufficient ignition energy necessary to achieve stable ignition of aluminum particles exposed to a shock pressure condition, and this is particularly true when the aluminized high explosive undergoes afterburning following a primary detonation of its base charge.

For measuring the relevant ignition temperature of aluminum particle or presumed its surface temperature at ignition, the experiments are quite challenging since a very short time is involved in the heating, ejection of particles, and the life time of initial plasma. Instead, we make an estimation of the ignition temperature based on the semi-infinite surface analysis of a heat diffusion process.
\[ \frac{dT}{dt} = \alpha \frac{d^2T}{dx^2}, \]  

(3)

where \( x \) is a depth of aluminum surface and \( \alpha \) is diffusivity. \( q'_0 \) is absorbed laser beam energy which has 4% absorptivity. During the ablation of a heated aluminum wire, the ignition temperature would be equivalent to the surface temperature of a wire. Assuming constant surface heat flux with initial temperature, the temperature distribution can be analytically obtained as

\[ T(x, t) - T_0 = \frac{2q'_0(\alpha t/\pi)^{1/2}}{k} \exp \left( -\frac{x^2}{4\alpha t} \right) \]

\[ - \frac{q'_0}{k} x \text{erfc} \left( \frac{x}{2\sqrt{\alpha t}} \right), \]

(4)

where \( T \) is ignition temperature, \( k \) is conductivity of aluminum (210 W/m K), and \( t \) is irradiation time (~500 \mu s). Because ignition occurs at the surface (i.e., \( x = 0 \)), Eq. (4) reduces to

\[ T(0, t) = T_0 + \frac{2q'_0(\alpha t/\pi)^{1/2}}{k} \]

(5)

and is plotted in Fig. 12.

The ignition temperature as shown is 975 K at 0.3 GPa. In atmospheric pressure, ignition temperature of 80 nm and 3 \( \mu \)m aluminum particles are 830 K and 950 K, respectively.44,45 However, ignition temperature at 2.2 GPa is 2150 K, which is twice the value at 0.3 GPa. This increase of ignition temperature is presumed characteristics of aluminum particles at a shock pressure condition.

Now to determine the effect of a pressure in the burning characteristics of aluminum particles, we measured the burning time by tracing the AIO band as previously explained. Figure 13(b) shows burning time vs. pressure for aluminum particles at 2000 K. Figure 13(a) is a reference data, showing the burning time at low pressure.5 In Ref. 6, the burning time for 80 nm particles decreased with increasing pressure and temperature, while for temperature above 2000 K, it remained constant regardless. The considered range of pressure was rather low at 0.8 and 3.2 MPa. Their results suggest that the burning time sensitivity to pressure and temperature is less significant at high temperature conditions above 2000 K.

In the present experiment, we considered the particles of 80 nm and 3 \( \mu \)m in size at \( \sim 2000 \) K and 0.3–2 GPa conditions and measured the relevant burning time. As illustrated in Fig. 13, the burning time for higher pressure range is reported to remain within 400 and 500 \mu s range. Making note of typical burning time for 3 \( \mu \)m aluminum particle being 500–600 \mu s,37,44 the result in Fig. 13(b) is quite consistent.

Now, the burning response of aluminum cloud containing either nano- or micro meter sized particles, exposed to a detonation product environment, is considered, suggesting that (1) more energy (determined) for ignition is required and (2) burning time at such condition will remain essentially constant. These are important characteristics of an explosion of the fuel-rich mixture either under-oxidized or heavily metalized that undergoes an afterburning phase, following a primary detonation that gives rise to an elevated extension of the initial wave.6

FIG. 10. Extended time images of Fig. 9(c) up to 719 \mu s.

FIG. 11. Ignition curve for aluminum particle at high pressure condition.

FIG. 12. Calculated ignition temperature from radiant temperature measurements plotted together with Ref. 44 data.
temperature and pressure condition similarly reproduced by the present laser experimental work.

IV. CONCLUSION

A laser-based experiment for igniting aluminum particles at shock pressure conditions up to 2.2 GPa has been conducted. The shock pressure induced by laser ablation resembles the detonating pressure of a high explosive (1–2 GPa), which provides an initial condition for a subsequent afterburning of the aluminum particles. The present experiment described how the shock pressure affects the ignition temperature and the burning time. The aluminum cloud generation from the ablative mass ejection was confirmed by a visualization scheme. The CO2 laser beam was used to supply necessary radiant thermal energy needed to ignite the ejected aluminum particles in air.

Based on this experiment, we found that first, radiant ignition temperature exhibits a log profile with respect to pressure, suggesting that more energy is required for ignition at higher pressure condition. This increasing ignition temperature with increasing pressure is presumed unique characteristics of aluminum particles. Second, the calculated ignition temperature at relatively lower pressure (0.3 GPa) is approximately 900 K, which is a known value for the nano-sized aluminum particles. Whereas the ignition temperature at an elevated pressure, for instance, 2 GPa, is approximately 2000 K which is about twice higher. Third, burning time of aluminum particles of sizes 80 nm and 3 μm varies just slightly between 400 and 500 μs for an entire range of pressures (0.3–2.2 GPa), suggesting that pressure has only a minor effect in the burning time. Instead, the burning time is directly related to a reaction time such that a short burning time implies a shorter reaction time. For a temperature ranging between 1200 and 2000 K, the reaction time decreases with increasing pressure. For temperature above 2000 K, this pressure dependence is insignificant such that the reaction time remains approximately constant. These findings on the burning time dependence on pressure for varying temperature provide an important range of pressure and temperature when building the reactive flow model of aluminum particles at high pressure conditions. Further tests are desired for examining the response of the larger particles, which is expected to be dependent on pressure.

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