High pressure study of aluminum particle ignition using lasers

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1 Introduction

Aluminum (Al) powders are commonly added during high explosive (HE) synthesis to modify the responsive pressure impulse and temperature profile of otherwise well-characterized detonation behavior of an HE. The time scale of energy release for reaction of Al particles is typically several orders of magnitude slower than that for the HE[1]. 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. Because high pressure tests up to a few GPa range is very limited, adding a dimension of high pressure dependence to the currently available predictive capability to the aluminized high explosive response at high pressure environment would thus strengthen the fidelity and applicability of a thermo-chemical model for such energetic materials of great interest. In a recent effort by Marion et al.[2], they measured the pressure dependence of the 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 GPa.

In this paper, our aim is to investigate the ignition characteristics of pure aluminum particles exposed to an extreme range of 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 aluminum particles in an air with pressure approaching those in the early expansion of a detonating explosive. First, the generation and dispersion event of particles are visualized to analyze the behavior of particle plume, and then the induced ablation is analyzed to predict the resulting pressure range. Second, the life time of plasma induced by a pulse laser alone is compared with a life time of flame induced by both pulse laser and continuous wave CO_2 laser. The plume is ignited by a CO_2 laser, and the emission spectroscopy is used to confirm the ignition and combustion of aluminum particle plume. The CO_2 laser energy provides heat source or radiant temperature for the particle ignition. The present laser-based ignition approach offers a novel control over the pressure condition, and in particular the CO_2

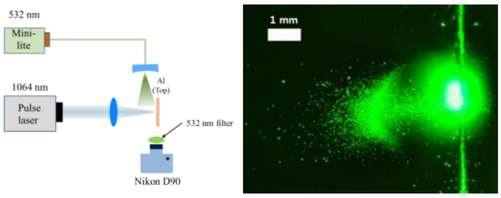
beam provides the desired thermal environment for aluminum ignition. The established ignition regime for laser irradiance at severely high pressure conditions of this work will be used when modeling a full detonation of an aluminized high explosive that consists of a strong afterburning following a main charge detonation.

2 Experimental methods

2.1 Generation of aluminum particles at severe pressure environment

Laser ablation technique has been proven effective for generating micro/nano-size particles [3] at a high pressure environment[4]. Particles generated from laser ablation have a narrow particle size distribution with variable primary particle size and shape. The flexibility of the setup further allows laser ablation for subsequent particle generation to be performed in air as well as in the liquid.

A procedure for particle generation via the laser ablation is as follows. Laser is irradiated on an aluminum surface, and small amount of aluminum is ablated due to phase explosion of the 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.



(a) (b)

Figure 1. A visualization scheme of micro/nano-aluminum particle generation and ejection (a) and the image of ejected aluminum particles at 25 μs after 100 mJ pulsed irradiation (b).

Aluminum is ablated by 1064 nm Nd:YAG pulse laser as shown in Fig. 1(a). The spot diameter and pulse duration are 1 mm and 9 ns, particles are ejected from a molten crater, and a 532 nm pulse laser with time delay is irradiated on the ejected particles for visualization (Fig. 1(b)). Light is scattered by the lifted particle plume and is collected by a camera [3]. The particle ejection image is captured at 25 μs after the incident 100 mJ irradiation. Target is an aluminum plate of 5x5x0.5 mm in size. The particle dispersion is vertical to an irradiated surface in the image. For estimating the ablation pressure of aluminum, a simple and useful relation in [5] is used between pressure *P* and laser irradiance *I*, such that

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

A is the atomic mass 26.98 and *Z* the average degree of ionization 220. The range of irradiance is 4~32 GW/cm^2 with a spot size of 1 mm diameter as this induced the pressure ranging between 0.35 and 2.2 GPa. The generated particles plume and the ablation pressure establish a similar environment for aluminum particles exposed to a severe pressure and temperature condition as intended in this research. This helps us to focus on the characteristics of aluminum ignition at elevated pressure condition suitable for the afterburning of aluminum particles in a hot product gas ambience.

2.2 Igniting at high pressure

Laser ablation using a pulsed laser establishes a high pressure condition adequate for the study of aluminum ignition while a continuous CO_2 laser provides control over the desired high temperature environment. Figure 2 shows a schematic of the laser and detection system for studying the aluminum particle ignition under high pressure and high temperature conditions.

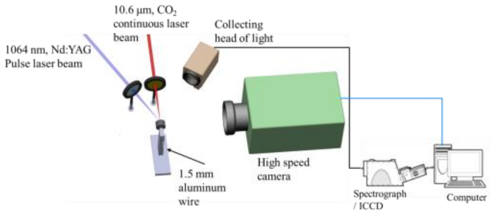


Figure 2. Schematic of ignition of aluminum particles under high pressure (via pulsed laser) and temperature environment (via continuous wave laser).

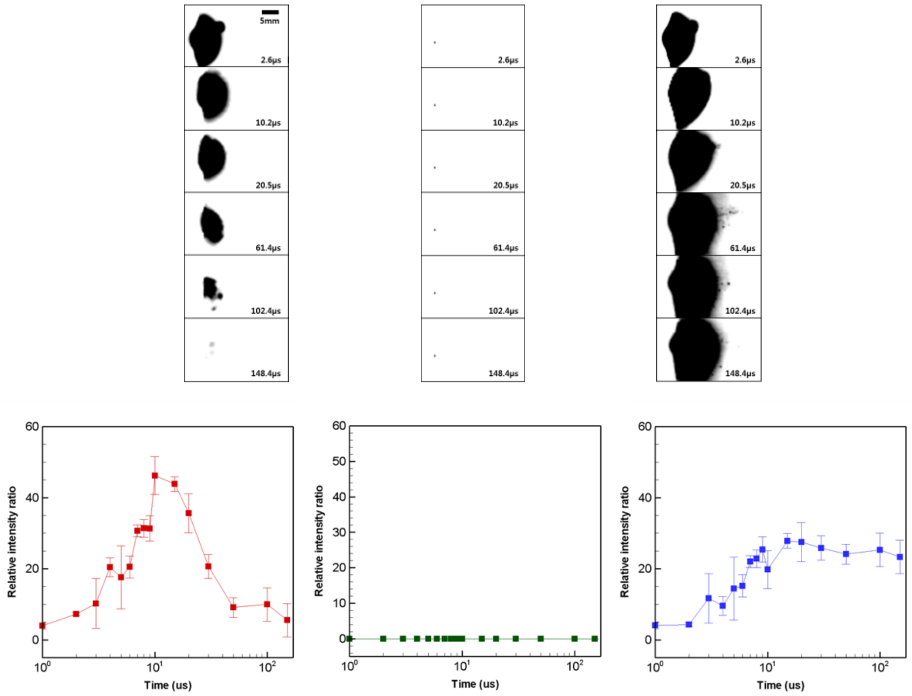
A CO_2 laser beam with 30~330 watts is irradiated on a 1 mm cross sectional area of an aluminum wire. The beam is aligned with the direction of particle ejection as such to maintain elevated temperature environment during the measurement. Then the temperature of radiant power is calculated by

$$q = \sigma T_r^{\ 4} \tag{2}$$

where q is the laser irradiance, σ is the Stefan-Boltzmann constant(5.67 x 10^{-8}W/m^{2}K^4) and T_r is the radiant temperature by laser. For the given laser irradiances, the resulting range of temperature is between 5000 and 9000 Kelvin. Plasma induced by pulse laser and flame of aluminum particles is visualized using high speed camera to compare the difference in motions. The ignition of aluminum particles is confirmed using spectroscopy analysis with tracing AlO band. Relative intensity of AlO vibronic wavelength(484 nm) is measured to track ignition.

3 Results

3.1 Ignition of aluminum particles



(a) Pulsed laser only (b) CO_2 laser only (c) Pulsed laser with CO_2 laser

Figure 3. The b/w inverted emission images of ignition (Upper) and measured relative intensity ratio of AlO band-484 nm (Lower) for each system setup: (a) Pulsed laser only, (b) CO_2 laser only, (c) Combined pulsed laser and CO_2 laser.

Visualization of the ignition helps to confirm appearance of aluminum particle flame and to measure and compare burning time and ignition delay. The irradiation from 260 mJ pulse laser alone (Fig. 3(a)) shows periods of induction and decay of the plasma. The plasma is relatively small and its strength diminished rapidly by the plasma cooling [3]. The plasma by a pulsed laser disappeared after 150 µs. 330 W CO_2 laser alone (Fig. 3(b)) shows only a small light spot and no burning at all while CO_2 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 CO_2 beam heating, Fig. 3(c) showed both plasma and burn. The initial plasma resembles case (a) of pulse laser alone. However, plasma and burn are sustained for duration exceeding 150 µs. Particles produced by laser ablation start to eject at 15 µs, and the particle ejection continues even after 150 µs. We trace 484 nm AlO vibronic wavelength using spectroscopy to confirm the ignition and burning of aluminum particles[6, 7]. The relative intensity ratio of AlO band (484 nm) in Fig. 3(a) rapidly decays beyond 10 µs when a pulse laser alone is used. However, the signal reaches its maximum at 11 µs and is continued when CO_2 beam is combined with pulsed irradiation in case (c). If the emission spectrum of case (c) was of 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 AlO 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. 3.

3.2 Ignition temperature of aluminum particles plume under high pressure

The equation of state (EOS) for aluminum reflects that the melting point of aluminum increases with pressure[8] 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 is obtained for varying pulse laser energy using Eq. (1) while radiant temperature of CO_2 laser is given by Eq. (2). CO_2 laser power reflects a desired radiant temperature for ignition in each calculated pressure condition, and then high speed camera images differentiate ignition from failure for the tests performed. Tests are repeated 10 times for constructing the error bar for each pressure condition. Figure 4 shows the resulting ignition boundary, plotted on a pressure vs. radiant temperature plane.

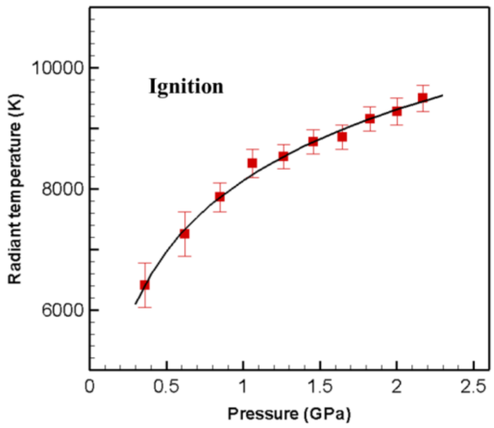


Figure 4. Ignition curve for aluminum particle at high pressure condition.

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

As 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 [9],

$$\frac{dT}{dt} = \alpha \frac{d^2T}{dx^2} \tag{3}$$

where x is a depth of aluminum surface and α is diffusivity. 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,

$$T(x,t) - T_0 = \frac{2q_0''(\alpha t/\pi)^{1/2}}{k} \exp\left(\frac{-x^2}{4\alpha t}\right) - \frac{q_0'' x}{k} \operatorname{erfc}\left(\frac{x}{2\sqrt{\alpha t}}\right) \tag{4}$$

where T is ignition temperature, k is conductivity of aluminum, and t is irradiation time. Because ignition occurs at the surface (i.e., x=0), Eq. (4) reduces to

$$T(0,t) = T_0 + \frac{2q_0''(\alpha t/\pi)^{1/2}}{k} \tag{5}$$

and plotted on Fig. 5.

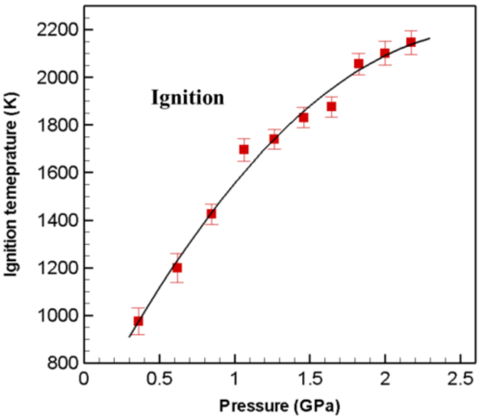


Figure 5. Dots are calculated from experimental results and black line is a fitting line.

The ignition temperature is shown 975 K at 0.3 GPa. This is in the proximity of experimentally known ignition temperature of a few hundred nano-meter sized particles [6]. 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 an elevated pressure condition.

4 Conclusions

A laser-based experiment for igniting aluminum particles at high temperature and pressure conditions has been conducted. The pressure induced by laser ablation resembles a detonation pressure of high explosive (1~2 GPa) which provides an initial ambient condition for a subsequent afterburning of aluminum powder of both nano- and micro-meter in size. The experiment was first aimed at describing how such high pressures affect the ignition temperature and burning time of ablated aluminum particle plume. The ablation pressure from the pulsed laser beam and the radiant temperature of the continuous CO_2 laser beam were estimated via empirical and theoretical means, respectively. The particle generation by laser ablation was confirmed by visualizing the ejecting particles. The CO_2 laser beam was used to supply necessary radiant thermal energy needed to ignite the ejected aluminum particles in air.

Based on this experiment, one discovers 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, 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.

Further tests at similar pressure conditions are desired for examining the kinetic behavior of the larger particles, which is expected to be dependent on pressure. The calculated ignition temperature and measured burning times of aluminum particles exposed to severe condition are expected to guide the authors in modeling of multiple reaction processes involving a heavily metalized high explosive. The presented ignition behavior of aluminum particles will improve the current state of predictive capability of non-ideal energetic material responses at large.

Acknowledgment

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