Ignition mechanisms of aluminum particles in hydrocarbon products

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1 Introduction

Aluminum plays a central role in the field of aluminized explosives, propulsion and energy production. The high energy density and chemical reactivity of aluminum particles significantly enhance the power of explosions. This enhancement is accompanied by an extension of the duration and intensity of the resulting pressures. However, the complexity of explosion phenomena involving aluminum particles poses a significant challenge to the study of the ignition of aluminum particles in fireballs. The main conditions that occurred during this explosion are detailed below.

Pressure in explosives can reach a high level (~100 kbar). As demonstrated by Kuhl et al. [1], most chemical reactions occur during the asymptotic phase (i.e., the final stage of the explosion). Due to thermal inertia and the high-pressure level in earlier phases, aluminum particles start combustion in this latest phase, when the pressure is about a hundred bars. In a related finding, Arafin et al. [2] determined that the melting temperature of alumina increases by a mere 2 K between 1 and 500 bars, suggesting that the melting temperature remains relatively constant. As will be discussed subsequently, the melting of alumina is imperative for the subsequent ignition of particles. The rate of heat release remains relatively stable for pressures below 100 bars, as noted by Alexandre Braconnier. Consequently, it can be preliminarily concluded that pressure does not exert a substantial influence on the combustion of aluminum particles.

A second criterion is the size of the particles. Most experimental works have been focused on the combustion of large (>10 µm) or individual particles or agglomerates [3-6]. In practical applications, aluminum burns as a dense cloud of micron-scale particles. Pokhil et al. [7] demonstrated that extrapolating combustion models from large particles to micron or submicron sizes can be misleading due to potential changes in the combustion regime. Sundaram et al. [8] showed that the ignition temperature is limited by the melting temperature of the oxide (2350 K) for large particles (>100 µm) and by the melting temperature of aluminum (933 K) for small

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particles (<1 µm). For the particles utilized in this study (7 µm), the ignition temperature considered is the melting temperature of the oxide.

During the asymptotic phase, the detonation products react with the oxygen of ambient air, creating a gaseous atmosphere composed of CO_2, H_2O, CO and O_2, in which aluminum particles react. Important convective transfers take place between metal particles and the hot atmosphere.

Aluminum can represent 20 to 30 % of the solid heterogeneous explosive mass. During the asymptotic phase, the particles are dispersed, leading to locally high concentrations. Some investigations of stabilized hybrid flames were performed to investigate aluminum burning in the burnt gases of methane-air mixtures (i.e., mixture of CO_2, H_2O and CO_2, N_2) [6,9]. They show that aluminum concentration has a significant influence on the combustion behavior of the hybrid mixture. Diffusive, convective and radiative transfers can be altered by a particle burning in a particle cloud.

In this study, a Bunsen-like burner operating at atmospheric pressure is utilized. The focal point of this investigation lies in the examination of the ignition and combustion mechanisms of aluminum particles within a stabilized flame of aluminum-seeded methane-oxygen-argon hybrid mixtures. The composition of the gaseous atmosphere in which the particles react is crucial for understanding these mechanisms. In the configuration employed, a methane (CH_4) and oxygen (O_2) gas mixture fosters a homogeneous and stable flame, thereby enabling the efficient heating and ignition of aluminum particles. The alteration of mixture composition affects the burnt gas temperature (hereafter called T_{CH_4/O_2}), which, in turn, modifies the convective effects between the burnt gases and particles. Figure 1 provides a visual representation of the interactions among various species and temperatures at different flame locations.

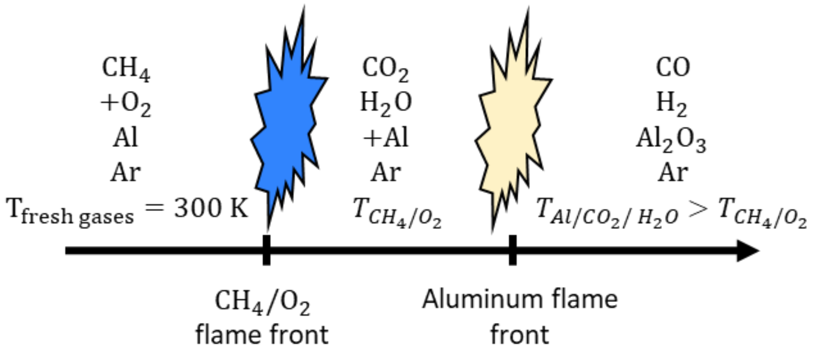

Figure 1: Scheme of a hybrid mixture fronts flame

In this study, two parameters were experimentally varied while maintaining the oxidizer composition constant: 1) the temperature of the surrounding atmosphere (T_{CH_4/O_2}), which directly impacts convective transfer, and 2) the concentration of aluminum particles, which affects radiative transfer between particles.

Optical measurements were conducted to identify the conditions necessary for the ignition of aluminum particles. Spectroscopic measurements were then leveraged to detect ignition indicators and estimate the flame temperature in the condensed phase. Synchronizing the measurement of particle concentration with emission spectroscopy enabled a direct correlation between flame temperature, aluminum mass, and the detection of intermediate species (Al, AlO). The initial findings of this investigation revealed a correlation between flame temperature and the particle concentration required to ignite an aluminum flame. Furthermore, a model of particle heating in a hydrocarbon flame is proposed to explain the ignition phenomenon of aluminum particles and provides an initial estimation of the radiative power needed to ignite a particle.

2 Experimental details

A. Dust Burner

The burner used in this work was initially developed for aluminum-air mixture experiments [10]. In this research, a hybrid methane-aluminum mixture is examined. A new dispersion system, allowing a homogeneous dispersion of micron-sized particles, has been implemented in a previous work [11]. Particles, stored in a sealed container, are transported by argon and premixed with other fresh gases. The initially turbulent flow is expanded and laminarized in a divergent pipe before exiting through a conical nozzle with a 2 cm inner diameter. A coflow surrounds the nozzle to create a coaxial flow of inert gases (argon) at the same speed as the mixture. The bulk flow is regulated by four flowmeters, allowing adjustments to the mixture composition and particle concentration. Two flowmeters control the argon flow rate: one for direct argon injection and the other for aluminum particles transport. Figure 2 describes the burner and the dispersion system.

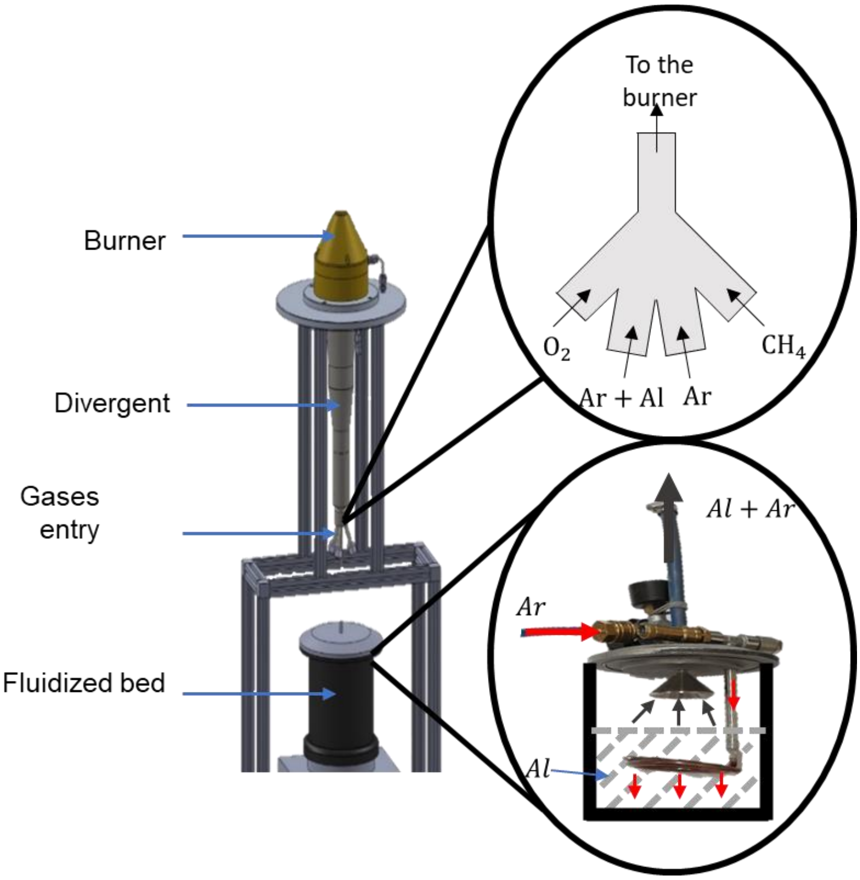

Figure 2: Scheme of the burner and dispersion system

B. Particles Concentration Measurements

The concentration of aluminum particles in the flow is continuously measured by a laser attenuation probe. This system consists of a diode laser ($\lambda = 632\text{nm}$) and two photodiode sensors. The laser emits a beam passing through the dusty flow to one of the diode sensors. This sensor records the transmitted beam and the flame emission, while the second one, used as a reference, captures only the flame signal. The particles concentration probe has been calibrated by comparing the total mass dispersed, measured by a weight placed under the burner, and the laser attenuation.

C. Flame Photography and Spectroscopy

A digital camera equipped with a neutral density filter is used to capture images of the flame. The spectral emission of the flame is acquired by a spectrometer aimed at the tip of the flame, where aluminum particles typically ignite first. The spectral emission is captured by a collimator connected to the spectroscope via a fiber-optic cable. Both the camera and the spectroscope are synchronized with the particles concentration measurement to be correlated with the instantaneous aluminum mass.

Spectroscopy is used to detect aluminum monoxide (AlO) to confirm the ignition of aluminum particles. Additionally, emission spectra are used to determine the temperature of the condensed phase by fitting the continuous part of the spectra to Planck's law of blackbody radiation, as described in [12,13].

D. Methodology

In this study, the ambient gas temperature is calculated using a theoretical equilibrium calculation for a stoichiometric methane-oxygen-argon flame. Table 1 presents the conditions tested in this work.

Table 1: Condition of mixture and temperature

O_2 molar fraction	CH_4 molar fraction	Ar molar fraction	Temperature [K]
0,163	0,081	0,756	2350
0,159	0,079	0,762	2325
0,155	0,077	0,768	2301
0,151	0,076	0,773	2275
0,148	0,074	0,779	2251
0,144	0,072	0,784	2225
0,141	0,070	0,789	2203

Once the conical gas-phase flame is stabilized over the nozzle, the particle concentration is gradually increased. To maintain a consistent temperature, the total argon flow rate is balanced: the flow rate through the sealed bowl containing aluminum particles is increased, while the direct argon injection is decreased. For each step of concentration, several spectra are recorded.

3 Results

Flame emission spectra are acquired at each concentration step. Spectra are used to detect the reaction intermediate of aluminum combustion, such as $Al_{(g)}$ and AlO. Figure 3 illustrates the evolution of concentration over time, with the methane-oxygen flame temperature estimated at 2325 K. Each vertical line corresponds to a moment when a spectrum and an image of the flame are acquired. As can be seen in figure 4, the colour and brightness of the flames vary depending on the aluminum concentration. Figure 5 regroups the recorded spectra.

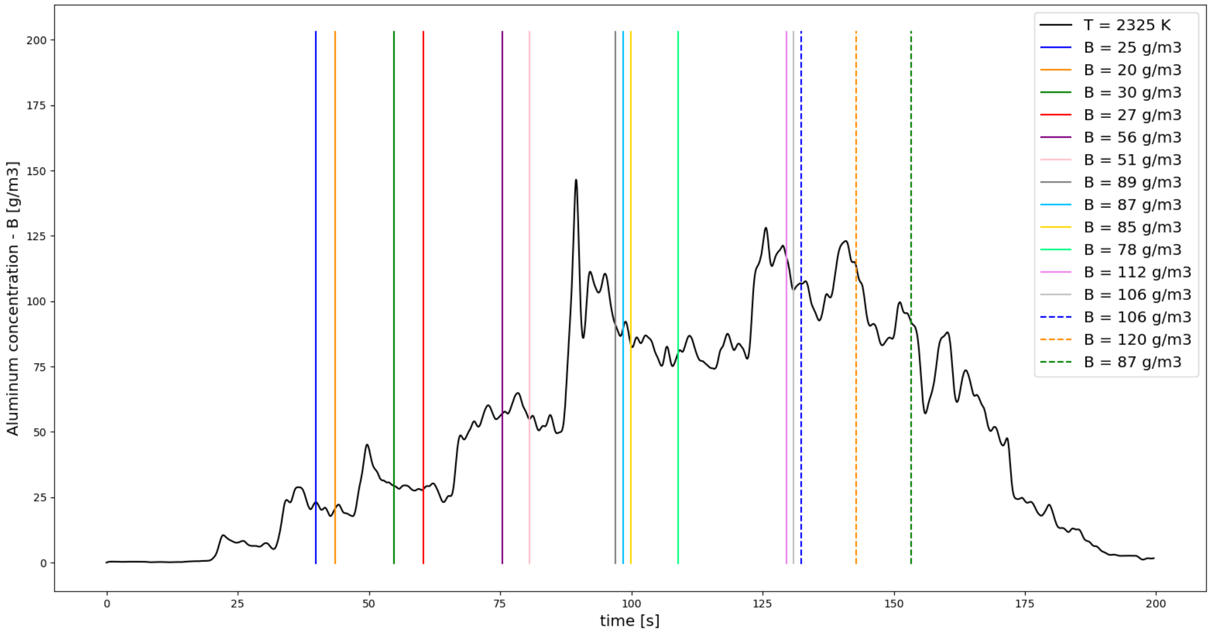

Figure 3: Evolution of the aluminum concentration over time

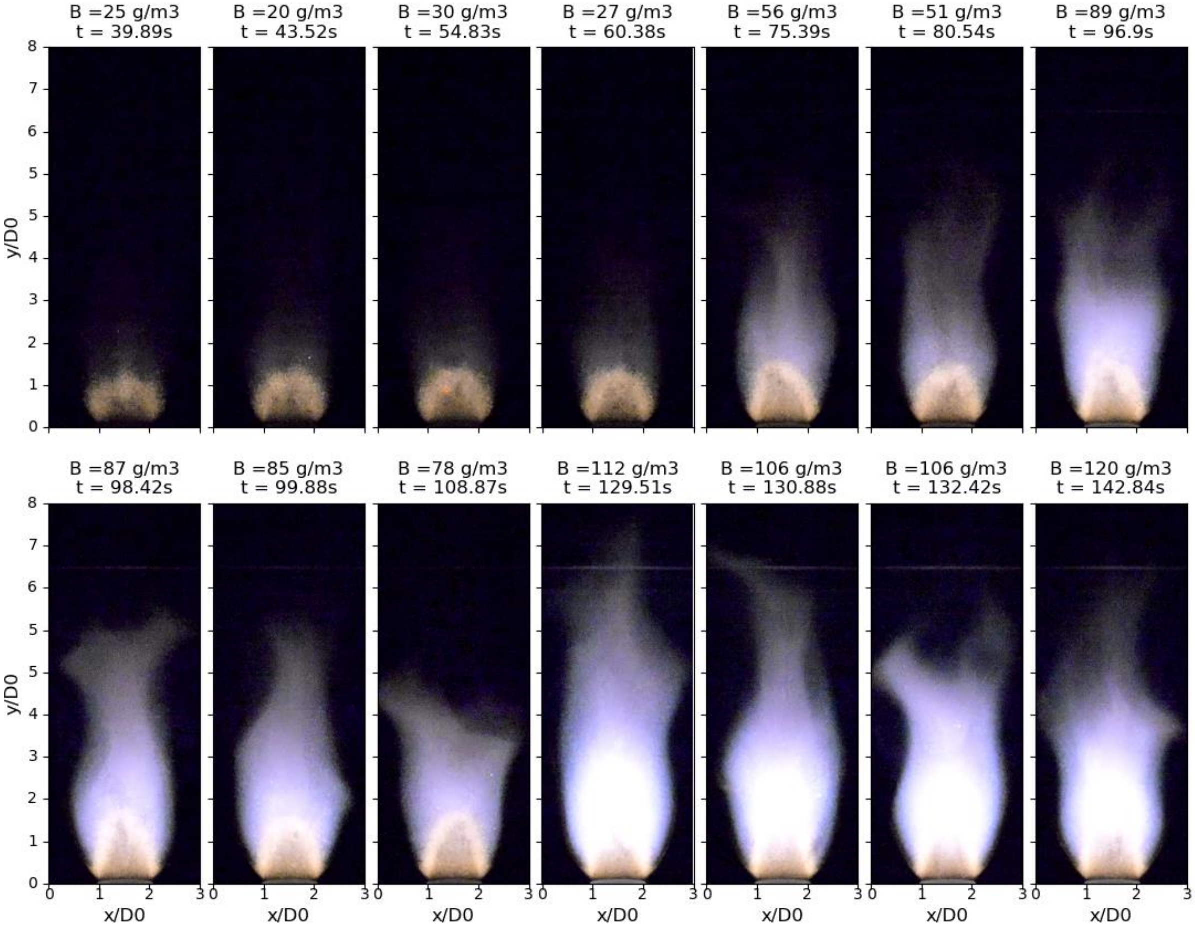

Figure 4: Visualization of flames at different concentration

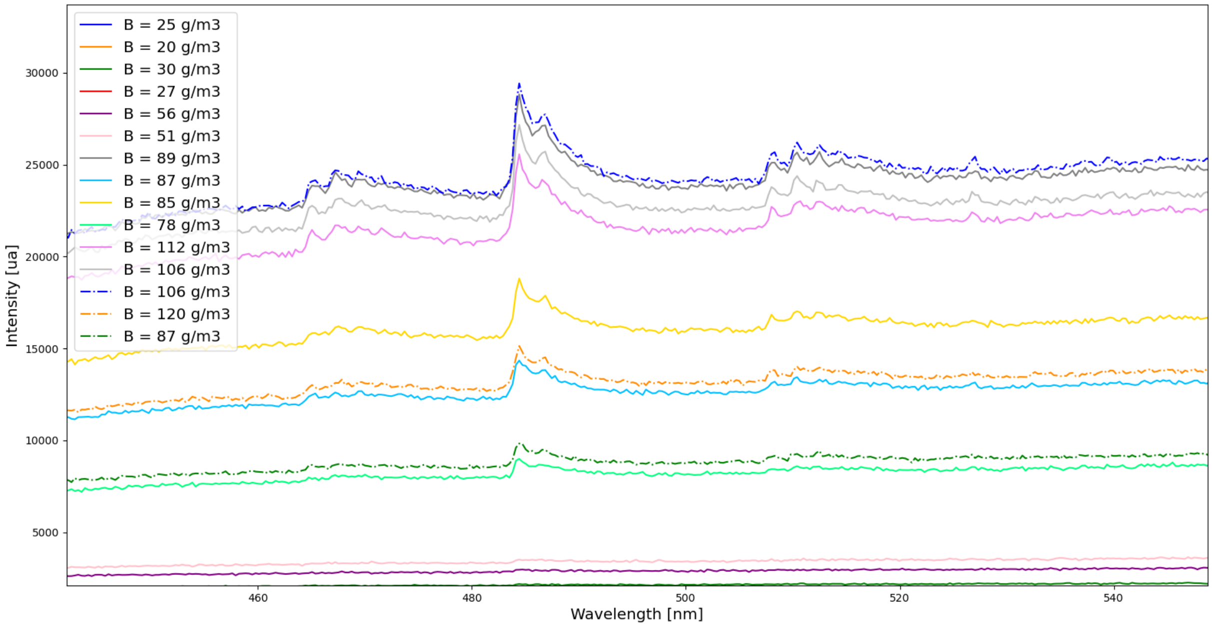

Figure 5: Flame emission spectra recorded at different concentration

The flame photographs in Figure 4 show that the flame structure evolves with varying concentrations. It appears that the ignition of aluminum particles starts from the tip of the conical flame. At low concentration, flames are yellow-orange and low brightness, indicating the heating of particles without combustion. As the concentration increases, the flame becomes progressively brighter and whiter. Additionally, the bluish hue of the plume can be attributed to the emission of the AlO main band at 485nm.

At low concentrations, the flame spectra mainly consist of continuous blackbody radiation. The AlO characteristic bands only appear when the concentration increases. These bands are not well developed at lower temperatures (only two characteristic bands), whereas at higher temperatures, all three bands are well pronounced. This test suggests that the ignition of aluminum particles begins at a concentration of around 89 g/m³. Figure 6 compiles the results of various tests, showing the threshold concentration above which aluminum ignites as a function of the burnt gas temperature. In other words, these are the minimum concentrations required to ignite aluminum particles.

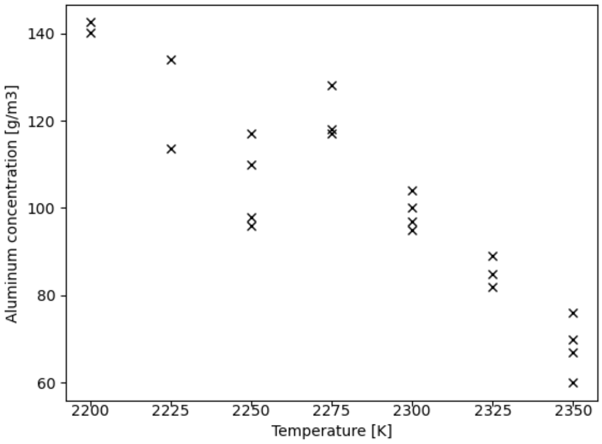

Figure 6: Concentration measured to ignite aluminum particles over ambient temperature

The minimum concentration required to ignite aluminum particles decreases as the temperature of the flue gases increases. In all these tests, the flue gas temperature remained below the melting point of alumina (2350 K). These results clearly demonstrate the role of radiative exchange in the heating and ignition of the particles.

5. Conclusion

The ignition of aluminum particles within the combustion products of a methane-oxygen-argon flame is influenced by both the ambient temperature and the concentration of aluminum. The observed decrease in the concentration required to ignite aluminum indicates that convective effects alone are insufficient to initiate combustion, suggesting that particles collectively contribute through radiative transfers. Future research will explore the boundaries of this phenomenon at varying temperatures. Additionally, a heating model will be developed to estimate the radiative transfers necessary to ignite a single particle within a dusty cloud.

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