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Experimental investigation of the aluminum combustion in SRM representative gas species: effect of CO₂ / CO

A. Braconnier, C. Chauveau**, F. Halter** and S. Gallier**

** ArianeGroup*

CRB, 9 rue Lavoisier, 91710 Vert-Le-Petit, France

*** ICARE-CNRS*

1C avenue de la recherche scientifique 45071 Orléans Cedex, France

Abstract

This work aims at providing a fundamental description of the aluminum combustion process in SRM representative gases, focusing on the phenomenology of a burning droplet reacting in pure CO₂ and CO by varying pressure. Using an electrostatic levitator, single micron-sized aluminum particles are isolated in a static gaseous environment to observe their self-sustained combustion with optical diagnostics. Combustion occurs in a diffusion mode in CO₂ and is limited to surface reactions in CO but, in each case, the combustion process seems driven by some condensed compounds accumulation on the droplet surface during reaction and modulated by the pressure.

1. Introduction

Solid rocket motors (SRM) remain a robust and reliable propulsion system selected to provide a valuable trade-off between performances and costs with a simple design. In order to increase combustion temperatures and motor-specific impulse, current composite propellants formulations contain aluminum powder as energetic additive. Thus, micron-sized aluminum particles react inside the SRM chamber with oxidizing species produced by the solid propellant combustion such as HCl, H₂O, CO and CO₂.

Ignition of aluminum wires in carbon dioxide was firstly observed in an experimental investigation [1]. Then, different dedicated studies were conducted to accurately understand the effect of CO₂ on the aluminum combustion process. Ignition conditions in terms of temperature and pressure were analyzed [2], showing that combustion of aluminum in carbon dioxide can occur in a diffusion mode with ignition temperature lower than the aluminum oxide (Al₂O₃) melting point (~2300 K). A primary kinetic model was proposed, presenting different gas phase reactions between Al components and CO₂ to produce CO and condensed Al₂O₃. Surface reactions were also integrated considering that CO₂ do not reach the droplet surface and only intermediate products AlO can react with the liquid aluminum [2]. Flame and droplet characteristics during combustion in CO₂ atmosphere were detailed to improve the understanding of aluminum combustion mechanisms [3-6]. Flame temperature is close to 3200 K, below the Al₂O₃ boiling point. Stoichiometric proportions of Al and O are found in the flame zone without trace of carbon, resulting in the formation of liquid aluminum oxide [3, 6]. Nano-particles layer is found on the particle surface and is also free of carbon, however, droplet is composed of different phases including Al, C and O compounds. These observations suggest that thermophoresis from the vapor-phase and heterogeneous reactions in the droplet can significantly affect combustion parameters. Data on combustion parameters in carbon dioxide were presented by various authors [3, 6-13]. Combustion time, combustion rate and flame size were mainly determined and often compared to the results in oxygenated environment, deducing overall that CO₂ has a lower oxidizer efficiency than O₂. Experimental correlations were established to estimate the burning time of Al particles depending on the initial droplet diameter D_0 and ambient oxidizing conditions [8, 12-14]. The usual theory predicted that the combustion time is proportional to D_0^n with $n = 2$ in the case of vapor phase reaction, but, because of the partial coverage of the droplet surface by aluminum oxide during combustion, n is experimentally reported to be reduced. Nevertheless, despite the large available dataset, the estimation of the exponent n is representative of the substantial data scatter that results from the different experimental conditions ($0.3 < n < 2.5$). Thus, while the determination of the combustion parameters of Al particles in CO₂ atmosphere is an important issue for practical applications, a detailed description of the combustion in pure CO₂ has to be firstly provided as reference using an experimental approach which allows a fundamental analysis.

Aluminum reaction in carbon monoxide is less documented. Qualitative description of the combustion process in pure CO atmosphere reports a low luminosity trace and indicates that no diffusion flame is observed, which is consistent with the assumption of a kinetic-limited combustion mode in these conditions [3]. Complementary measurements of the species distribution also suggest that heterogeneous reactions possibly occur on the droplet surface, generating a complex condensed-phase near the burnt particles surface with Al₂O₃ and carbon-containing compounds. However, even if efficiency of CO as an oxidizer seems limited, it was noted that surface reactions can have a significant role in

the case of carbon-containing oxidizers like CO_2 [6]. According to these observations and reminding that the content of CO is significant in SRM combustion chamber, further investigations are required to understand the contribution of carbon monoxide in aluminum combustion and potentially state on its consideration in experimental correlations which actually do not integrate CO as an oxidizer [14].

Finally, in order to contribute to the ongoing research, the current work is focused on the qualitative characterization of the combustion process of aluminum particles in pure CO_2 and CO at various pressures. Particle diameters are selected to be representative of the particle sizes found close to solid propellant surface (30 to 120 μm , considering agglomeration). Single levitated particles are isolated in static and controlled gaseous environments and observed with high temporal and spatial resolution to finely evaluate their combustion processes. This study is dedicated to provide new fundamental information on aluminum combustion in SRM representative oxidizing environment to improve the phenomenology understanding which is required to develop accurate and reliable prediction models.

2. Experimental set-up

2.1 Apparatus

A specific apparatus was developed to fundamentally analyze the combustion process of isolated aluminum particles while limiting any contributions of the experimental conditions. Usual methods used to experiment aluminum combustion are limited to ensure a non-intrusive effect on the observed phenomena and can have an influence which potentially alters the different measurements. Indeed, in the case of shock tubes, free-falling droplet, aerodynamic levitators or bulk aluminum samples attached to support, many additional heat transfers have to be considered, respectively by radiation, convection and conduction. Based on elementary principles and designs [15], isolation of a single charged particle without any perturbing support is performed using an electric field to counteract gravity in an electrodynamic levitator (EDL). The EDL is composed of distinct electrodes supplied by AC and DC voltages which allow to set the horizontal and vertical position of the aluminum particle. Despite its influence on charged solid particles, the electric field is only used to set a reproducible initial droplet position. Indeed, considerable charge losses are assumed to occur at high temperature by thermoionic emission during combustion preserving condensed phase from any influence of the electric field. Several investigations were conducted on this issue and corroborate this assumption. Estimations of the thermoionic emissions demonstrate a rate of charge loss greater than that which can be handled by the EDL. The experimental set-up was also used to evaluate the free-falling speed of a heated aluminum droplet in a non-oxidizing environment to show that the falling speed corresponds to the value expected by the Stokes' law, proving that the effect of the electric field is negligible on condensate elements at high temperature. Moreover, despite possible ionization in the flame, axisymmetric geometry of the electric field inside the EDL preserves combustion from any preferential and perturbing electric effects on the reacting zone.

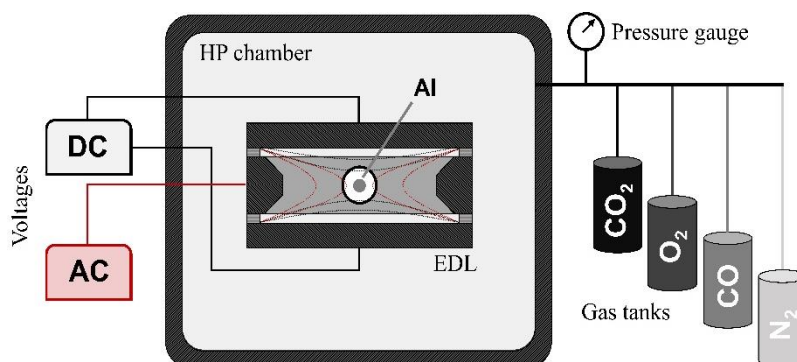


Figure 1 : Schematic representation of the experimental apparatus

This system is confined in a high pressure chamber to ensure a static and controlled gaseous environment. Partial pressures of the ambient mixture are adjusted with a high precision manometer. This experimental apparatus is schematically represented in Fig. 1. A 50 Watt CO_2 laser beam follows a defined optical path, being successively separated, reflected and focused to symmetrically irradiate and ignite the aluminum particle.

2.2 Diagnostics and measurements

Diagnostics mainly consist in optical measurements. Schematic representation of the entire experimental set-up and typical measurements are presented in Fig. 2. The light emission of the burning particle is collected and transmitted to filtered photomultiplier tubes (PM) used to transform the luminous intensity in recorded voltage signals with a rate up to 200 kHz (Fig. 2 (a)). Dynamic sequences of the combustion process are recorded with high temporal and spatial resolution (up to 40000 fps / 2.5 $\mu\text{m}/\text{px}$) using a high-speed camera PHANTOM V1611 combined to a long-distance microscope QUESTAR QM100 focused on the isolated droplet (Fig. 2 (b)).

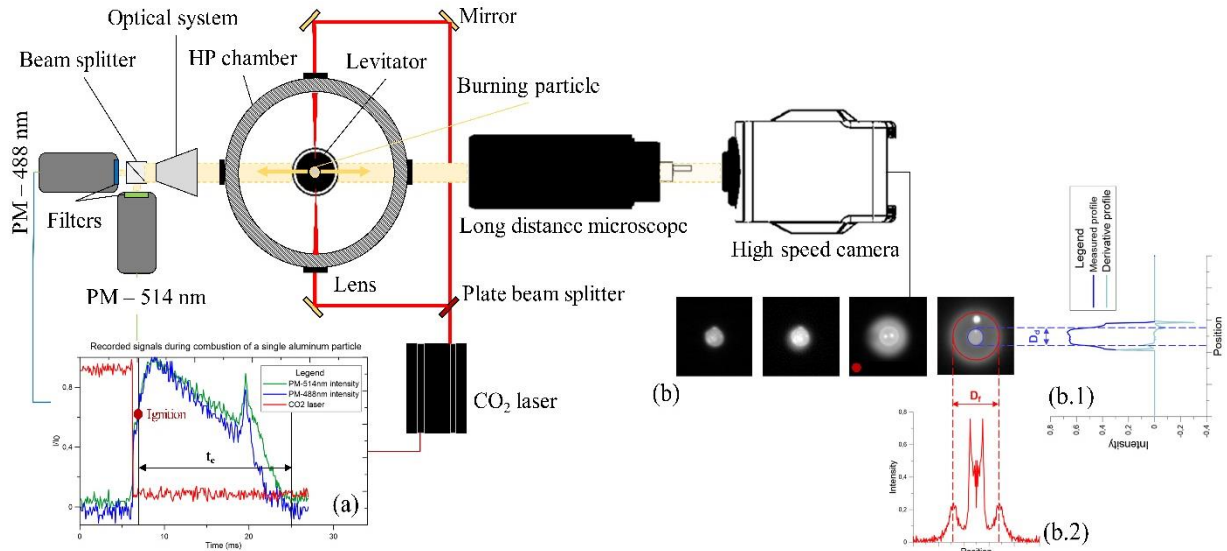


Figure 2: Schematic representation of the experimental setup and typical measurements

2.3 Protocol and processing

Ambient conditions are firstly fixed in terms of pressure and composition inside the high pressure chamber. Aluminum particles are then injected inside the EDL charged by triboelectric effect in the plastic injector. A single sample is hence isolated in the EDL by adjusting voltage parameters and finally ignited by the laser beam. The CO₂ laser is synchronized with photomultiplier to trigger its extinction when the luminous emission from the heated particle exceeds a defined threshold, ensuring the self-sustained combustion of the aluminum droplet (Fig. 2 (a)). The camera recording is also coupled to the laser trigger which is the temporal origin.

The different combustion parameters can be determined by using a home-made Matlab processing routine. Measurement of the initial droplet diameter and temporal evolution of the droplet/flame diameters is based on intensity profile analysis (Fig. 2 (b.1 – b.2)). Presented works will remain focused on describing the fundamental phenomenology of the aluminum combustion process but more detail on post-processing can be found in [16].

3. Results

3.1 Aluminum combustion in CO₂

The self-sustained aluminum combustion process in CO₂ atmosphere is illustrated in Fig. 3. Upper pictures are raw unprocessed and a Laplacian filter is applied for lower images. The second derivative is approximated in both directions of the picture and represented as a gray scale to allow contour detection. This image processing is used to highlight emissive structures (colored zones) and their evolution.

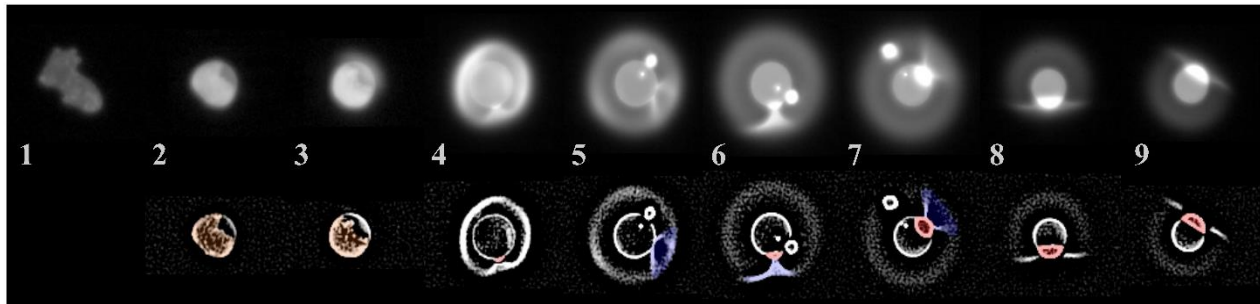


Figure 3: Raw and filtered pictures of the combustion process in pure CO₂ at atmospheric pressure

The signals recorded during the combustion sequence in Fig. 3 and the measured regression of the droplet diameter are introduced in Fig. 4 with corresponding temporal references.

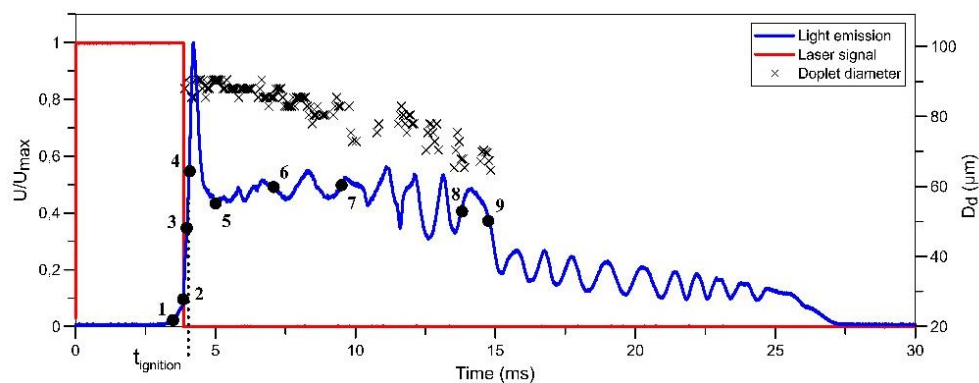


Figure 4: Temporal evolution of the recorded signals and droplet diameter during combustion process in pure CO₂ at atmospheric pressure. Numbers correspond to the images from Fig. 3.

Firstly, the solid aluminum particle is heated by the laser beam which increases the particle temperature and its subsequent light emission until ignition (Fig. 3-4 1-3). During this stage, the transition phase of the natural alumina layer which covers the aluminum particle is visualized. This initially solid oxide layer (Fig. 3 1) is observed as an intermediate phase colored in orange (Fig. 3 2-3) before it transits to a totally melted phase located on the aluminum droplet surface as a liquid oxide lobe colored in red (Fig. 3 4). A luminous flame can be unambiguously observed around the burning droplet after ignition with typical distinct gaseous and condensed phases (Fig. 3 4-9), supporting that combustion in pure CO₂ can occur in a self-sustained diffusion mode. Development of the bright flame is identified by an increasing light emission signal due to the important initial formation of condensate aluminum oxide in the nascent flame (Fig. 4 3-4). During combustion, oscillatory patterns are often detected on the photomultiplier signals due to the global spin of the burning particle and its oxide cap. The droplet and flame diameters decrease while the oxide lobe gradually grows up, partially covering the aluminum droplet (Fig. 3 4-9) and the overall luminous intensity progressively reduces (Fig. 4). The oxide cap blocked a part of the outward gaseous flow, generating a distinct zone (blue colored) over the covered reacting surface (Fig. 3 5-7). Then, an important part of the droplet surface being coated, the spherical flame becomes substantially asymmetric for a large part of the combustion process (Fig. 3 8-9). Nonetheless, the symmetric/asymmetric regime transition in the case of aluminum burning in CO₂ appears as more progressive and less violent than in O₂ atmospheres. Indeed, for aluminum particles burning in O₂ and O₂/N₂, transition from a symmetric to an asymmetric regime was found to be characterized by a significant luminous emission and considerable movements of the droplet (spinning / jetting) quickly followed by an abrupt extinction [16]. Considering the important duration of the burning time for aluminum particles reacting in pure CO₂, and the limited visualization field, extinction is never directly observed at atmospheric pressure although it is evidenced through the PM.

Fig. 5 illustrates a combustion sequence of an aluminum particle in pressurized CO₂ atmosphere ($P = 5$ bar) with raw and filtered images while the corresponding emission signal is introduced in Fig. 6. Pictures show that the flame appears to be brighter than at atmospheric pressure (Fig. 5 3-4). The formation of the initial alumina lobe (red colored) remains observed (Fig. 5 4) and the distribution of condensed alumina in the flame is heterogeneous forming emissive structures around the burning droplet (Fig. 5 4). Additional surrounding aluminum oxide is progressively evacuated outside but also contributes to quickly cover the droplet surface, forming a spreading layer (red colored). This phase corresponds to the first emission decrease (Fig. 6 3-4). An important part of the droplet is passivated by the oxide lobe and the combustion is hence altered. The flame becomes distinctly asymmetric (Fig. 5 5-6) and the luminous intensity reduces (Fig. 6 5-6). The relative surface covered by the liquid aluminum oxide layer continues to gradually increase,

the oxidizer supply is then inhibited, preventing from maintaining the combustion process and leading to an early extinction when the droplet is totally enclosed (Fig. 4 7-9) with the brief appearance of a bubble structure (Fig. 5 8). Finally, a considerable amount of aluminum oxide composes the combustion residue.

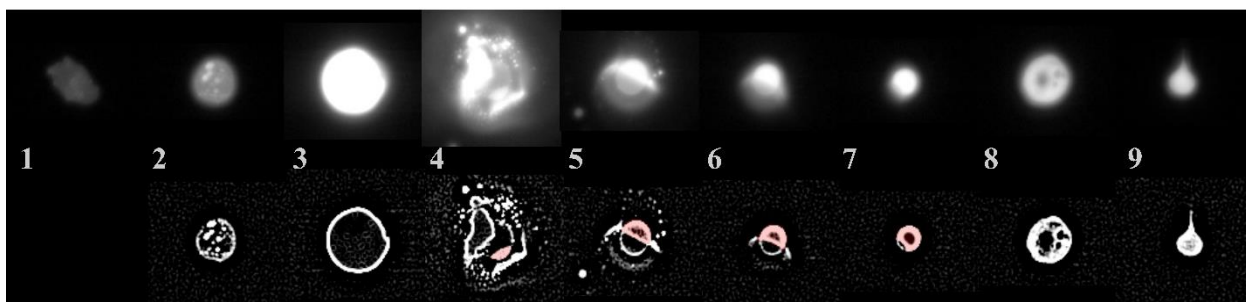


Figure 5: Video pictures of the combustion of an aluminum particle in pure CO₂ with 5 bar pressure

Photomultiplier signals presented in Fig. 6 for various pressures and similar initial droplet diameters ($D_0 \sim 73 \mu\text{m}$) indicate that the light emission evolution is analogous but emissions intensify by increasing pressure. Analysis of the emission traces also highlights the pressure effect on the combustion process. Temporal comparison between the luminous intensity and the pictures suggest that the first high emission level corresponds to the step where alumina structures are found in the flame zone (Fig. 6 3-4) Then, the signal drop marks the regime transition to a low emissive asymmetric phase (Fig. 4 5-7) which persists until extinction (Fig. 4 8-9). These different steps seem to constantly occur for higher pressure ranges ($> 1 \text{ bar}$) but are not identified for atmospheric pressures, both on image sequences or photomultiplier signals.

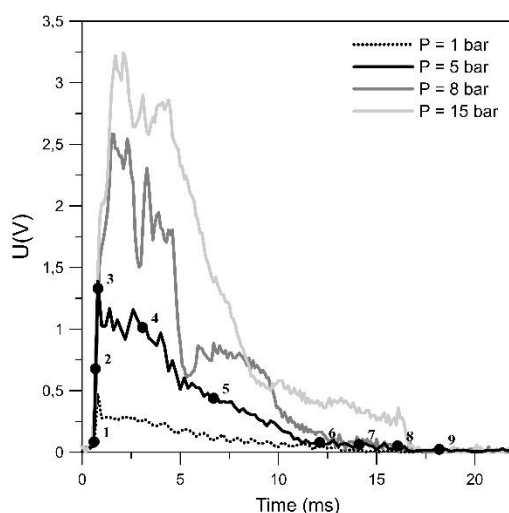


Figure 6: Emission signals of aluminum particles burning in pure CO₂ for varying pressure conditions.

3.2 Aluminum reaction in CO

Aluminum reaction in pure CO varying pressure was also investigated to provide information on the role of the carbon monoxide as an oxidizer. Different tests were also conducted in pure N₂ to allow a comparative analysis with a non-reactive environment as a reference. Pictures of aluminum particles heated in pure CO as well as typical emission signals for various pressure conditions in pure CO or N₂ are presented in Fig. 7 and Fig. 8 for a fixed laser extinction threshold and similar initial droplet diameters ($D_0 \sim 80 \mu\text{m}$).

In the case of aluminum particles heated in pure CO, video images show that the aluminum particle melts, forming a spherical aluminum droplet and a liquid oxide lobe (red colored) for all the tested pressure ranges (Fig. 7 2). This step corresponds to the initial rise of the luminous signals (Fig. 8 1-2). In an inert atmosphere, no reactions between aluminum and nitrogen are observed and photomultiplier signals immediately decrease down to their initial value after the laser extinction (Fig. 8). Emission signals of particles heated in CO are therefore different from that in N₂. Diverse trends are identified according to the pressure and images exposed in Fig. 8 corroborate that the phenomenology of the aluminum reaction in pure CO depends on the ambient conditions. The pressure is firstly found to have an effect on the luminous intensity and on the emission time for aluminum particles heated in carbon monoxide atmosphere. The

magnitude of the emitted light increases with pressure as well as the duration of the light radiation after the laser extinction. This influence is not noticed in nitrogen and pressure does not alter the light emission, either its intensity or its duration. Overall, those results are consistent with significant heterogeneous reactions between CO and aluminum surface. However, the energy release may not be sufficient to allow for a self-sustained combustion.

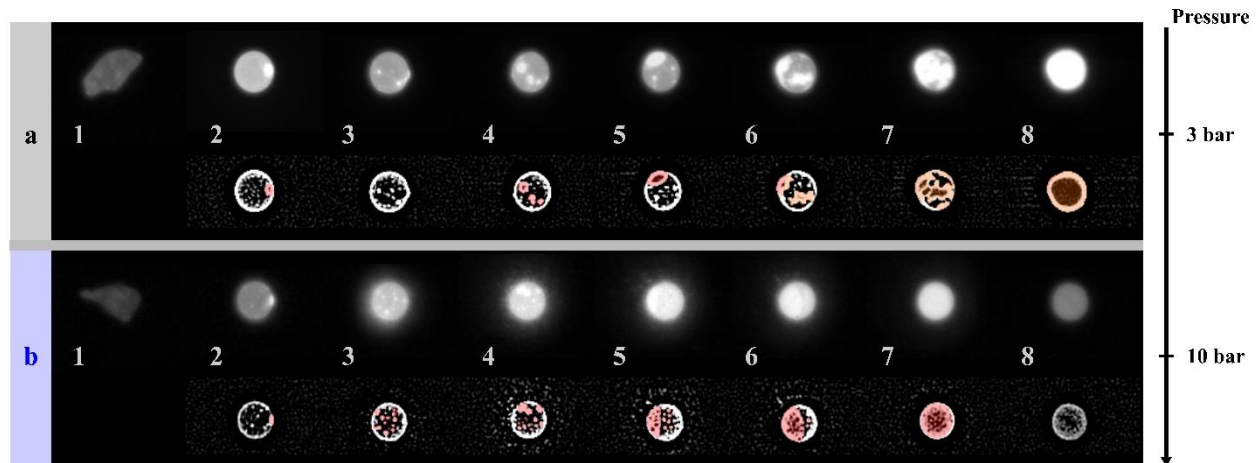


Figure 7: Video pictures of the reaction of aluminum particles in pure CO at 3 and 10 bar

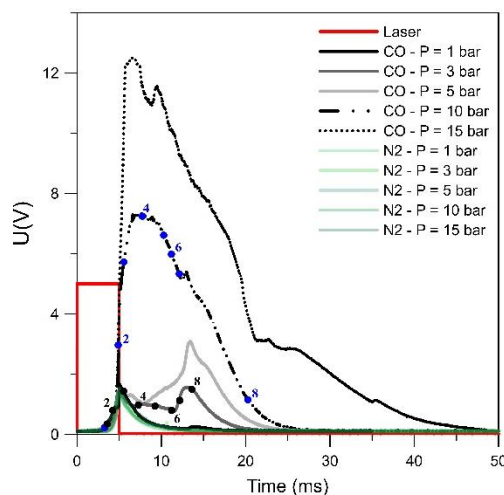


Figure 8: Emission signals of aluminum particles heated in CO or N₂ for various pressure conditions. Temporal references with the pictures in Fig. 8 for pressures of 3 bar (black spots) and 10 bar (blue spots)

In the case of lower pressure ranges, from 1 bar to 5 bar, the emission signals firstly decrease after the end of the heating phase (Fig. 8 3-4), following the typical luminous decay of an aluminum particle heated in a N₂ inert environment. Then, an evolution of the emission signal is observed (Fig. 8 4-5) corresponding to the production and aggregation of bright liquid spots (red colored) on the droplet surface (Fig. 7 4a-5a). The last step consists in the formation of a growing layer (orange colored) on the droplet surface which finally covers the entire particle (Fig. 7 6a-8a). This mechanism is notably identified by an emission peak before a reduction of the light intensity which gradually recovers its initial value (Fig. 8 4-6). The duration before the layer development occurrence t_{lr} compared to the total emission time after laser extinction t_e is presented on Fig. 9 for different pressures. This transition repeatedly occurs for fixed pressure and droplet diameter but appears significantly earlier when pressure rises.

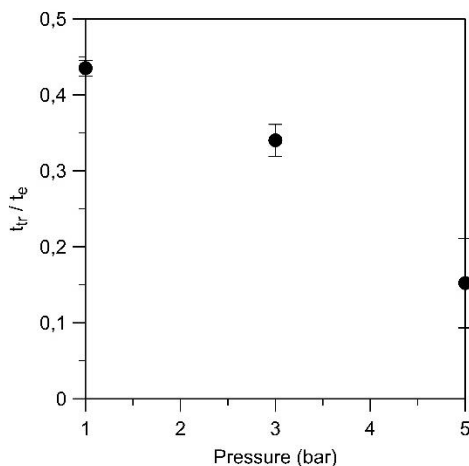


Figure 9: Relative duration before layer expansion occurrence for low and intermediate pressure ranges

For high pressure conditions (> 10 bar), the luminous intensity increases after the laser extinction (Fig. 8 2-3) and a surrounding bright halo can be discerned around the reacting particle (Fig. 7 3b-6b). Emissive liquid structures (red colored) are also formed on the droplet surface (Fig. 7 3b-5b), however, the sudden emergence of an emissive layer is not identified, the particle is finally progressively covered by the liquid products (Fig. 7 6b-7b) and the luminous intensity decreases (Fig. 8 5-8). Independently of the pressure conditions, an important quantity of aluminum is expected to remain unreacted after reaction in pure CO.

Laser extinction threshold was also varied to modify the duration of the heating phase and analyze the influence of the initial energy supply. Investigations report that a sufficient energy is required to initiate the different surface phenomena. Thus, when the droplet temperature is below the alumina melting point, the aluminum particle melts, the initial oxide lobe is not formed and the droplet emission rapidly decreases as it is observed in an inert environment. Moreover, it appears that pressure contributes to initiate reactions. Indeed, at elevated pressures, surface phenomena are identified although the initial additional energy is smaller. This is again consistent with heterogeneous kinetically-limited reactions.

4. Analysis and discussion

This part introduces qualitative analysis of the presented investigations to provide complementary understanding on the burning process of aluminum particles in CO_2 and CO. Results firstly demonstrated that the reaction of an aluminum particle in pure CO_2 and pure CO is significantly different for the studied droplet diameter range. The reaction in carbon dioxide is obviously identified as a combustion process in a diffusion mode, nonetheless, self-sustained reaction in vapor-phase was not observed in carbon monoxide after the energy supply extinction. However, comparison between particles heated in CO and inert environment proved that effective heterogeneous surface reactions are likely to occur in pure carbon monoxide. Reported kinetic studies [17, 18] can explain those observations, indicating that Al- CO_2 reaction can occur by different pathways (CO_2 dissociation or direct oxygen abstraction) whereas the strong CO energy bond would prevent from any gaseous Al-CO reaction.

As supposed [2] the combustion ignition in carbon dioxide can potentially occur before reaching the alumina melting point. Indeed, in Fig. 3, gas reaction is observed while the initial oxide layer is not molten yet. A liquid oxide lobe is then formed, remaining on the droplet surface and progressively growing. Aggregation of aluminum oxide on the particle surface can be attributed to various processes. Liquid oxide can be directly produced by surface reactions or condensed alumina can diffuse toward the droplet surface under temperature or concentration gradients. According to Fig. 3 and Fig. 5, aluminum surface seems homogeneous outside the oxide cap and no surface reactions are clearly identified in CO_2 atmospheres. Furthermore, no singular light emission is detected on the different photomultiplier signals in Fig. 6. This information implies that there is no spontaneous formation of emissive structures during the early phases of the combustion process, corroborating that the role of heterogeneous surface reactions on the oxide lobe supply seems restricted. Therefore, experimental assumptions imply that the oxide cap growing is mainly driven by thermophoresis or diffusiophoresis, corroborating reported expectations [6]. Moreover, the growing oxide lobe constitutes an obstacle for the outward gaseous aluminum flow and generates specific flow structures over the lobe (Fig. 3 4-7). This effect potentially induces the recirculation of submicronic condensed alumina particles from the flame to the droplet surface and can also promote the aggregation of liquid oxide. Although observations indicate that heterogeneous surface reactions seems inexistent or at least quite limited, the formation of complex Al-O-C phases was identified in quenched totally burned particles [6]. This suggests that production of Al-O-C compounds does not

occur during high temperature reactions as long as the diffusion regime is sustained. Thus, results are consistent with the supposed mechanism which predicts that phases transitions occur during particles cooling, forming Al-C and Al-O-C solutions at the end of the combustion process [6]. Based on this description of a burning droplet in pure CO₂, the D^2 law seems unlikely to be representative of the various observed phenomena. The reacting surface continuously reduces during the combustion process and the evaporation rate is expected to decrease near the extinction, that is why the consideration of a constant evaporation rate during burning phase is possibly not appropriate and considerable amount of aluminum is expected to remain unburned before extinction (Fig. 4 6-9). Finally, hypothesis of the classical D^2 are questionable and the oxide lobe influence has to be considered while integrating the pressure effect on its phenomenology.

Results introduced qualitative information detailing the reaction process of aluminum particles in pure carbon monoxide and suggested that the combustion regime results in heterogeneous surface mechanisms. The important CO energy bond considerably reduces the availability in oxygen atom and inhibits reactions in gas phase. Based on previous observations [3], stoichiometric ratio of Al and O was found near the droplet surface whereas no carbon compounds were identified at this location. Thus, the phase observed to be produced on the droplet surface is likely liquid Al₂O₃ and aggregation of the initial oxide lobe with formed liquid products corroborates that these phases possibly have similar compositions. The sudden droplet covering that appears during the reaction process at lower pressures seems to be due to the development and expansion of a possibly different phase. Indeed, this phase abruptly emerges from visually clean zones on the droplet surface, moreover, the liquid oxide previously accumulated remains observed on the droplet surface (Fig. 7 7a-8a), indicating that the phases are likely to be immiscible. This information suggests that the emerging phase is different from initial Al₂O₃ and probably results of a temperature phase transition during cooling particle. The extinction is hence supposed to occur because the reaction heat release is not sufficient to allow for a self-sustained combustion.

The exact evolution of the internal droplet phase changes cannot be proposed based on our results. Determination of the temperature history during the combustion processes as well as additional information about the temperature transitions that occurs in the Al-O-C system are needed to evaluate the product speciation. Some qualitative interpretations are founded on expectations about mutual miscibility of aluminum liquid phases, however, detailed information on this issue is also required to support a reliable analysis. The pressure effect on the combustion process both in CO₂ and CO mainly consist in altering the accumulation mechanism of liquid phases on the droplet surface. This parameter mainly modifies the aluminum phase changes temperatures and the oxidizers concentration. Thus, phase diagrams predicting potential liquid and gaseous reactions depending on temperature and species concentrations are also necessary to explain and understand the pressure influence on the burning reactions in carbon-containing oxidizers.

5. Conclusion

The aluminum burning process in pure CO₂ and CO with different pressures was analyzed using an advanced experimental approach. High spatio-temporal resolution video sequences completed by photomultiplier signals provide detailed descriptions of the fundamental phenomenology.

The combustion regimes in the studied oxidizers are identified to be significantly different. The CO₂ atmosphere ensures oxygen availability and aluminum combustion occurs in diffusion mode whereas the strong energy bond of CO prevents from any self-sustained vapor-phase mechanism and reactions are limited to heterogeneous surface reactions. But the energy release is not sufficient to provide a self-sustained combustion. However, results indicate that the accumulation of liquid phases on the droplet surface alter the burning process in both oxidizing environments. In pure CO₂, thermophoretic or diffusiophoretic flows are suspected to induce the retro-diffusion of condensed aluminum oxide produced in the flame zone toward the droplet. Apparent aerodynamic effects over the oxide lobe can also have a direct contribution on the liquid phase accumulation by creating recirculation zones. The aggregation mechanism of condensed phase on the droplet surface is different for aluminum reacting in pure CO. Liquid products are directly produced by heterogeneous surface reactions and possible phase transitions to form an expanding passivating layer. In each condition, the pressure also has an influence on combustion and the accumulation process seems to be subsequently altered. Early extinction occurs in carbon dioxide as well as in carbon monoxide. Detailed observations show that a relatively important quantity of aluminum remained unburned at the end of the combustion reaction, especially in pure CO. Significant regression of the droplet diameter in CO₂ has been determined but the growing oxide lobe is observed to gradually fully cover the particle surface, preventing from a total reaction of the aluminum. Use of the D^2 law to predict combustion parameters of a droplet burning in CO₂ seems hence unlikely to be pertinent. The droplet diameter of a particle reacting in CO does not significantly reduce and the reaction is finally terminated either when the energetic balance does not allow to maintain sufficient temperatures which sustain surface reactions or when aluminum droplet surface is totally covered.

An attempt to describe the exact composition of the visually observed phases is limited using our observations. Evolution of the internal droplet phase appears to be limited during the burning phase in CO₂ and potential Al-O-C phases are supposed to be produced near the extinction by temperature phase transitions. Condensed Al₂O₃ is assumed to be continuously produced on the droplet surface during reaction in CO before the formation of a different phase during particle cooling. Qualitative hypothesis was assumed based on previous reported works and interpretation of mutual miscibility. Finally, complementary measurements on temperature history and speciation during reaction as well as additional data on temperature changes in Al-O-C system are required to fully understand the combustion phenomenology and develop representative models.

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