



Aluminum combustion in wet and dry CO₂: Consequences for surface reactions

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Abstract

The combustion of aluminum droplets in wet (3 mol% H₂O) and dry CO₂ is studied in order to identify the influence of the two atmospheres on the surface processes. Millimeter-sized samples are maintained contactless in an aerodynamic levitation system and are heated continuously during burning by a laser. Ignition and combustion of the aluminum droplet are observed with a high-speed camera, the Al surface temperature is measured by an optical pyrometer, and unburnt residues are analyzed by X-ray diffraction. The determination of the burning rates and of the droplet temperatures reveals no differences between wet and dry CO₂ ($\beta = 1.28 \pm 0.05 \text{ mm}^2/\text{s}$, $T = 2600 \pm 50 \text{ K}$), which shows that the gas-phase combustion regime is not affected by the presence of water vapor. However, the oxide cap, initially formed by the oxide coating breakdown at ignition, is progressively removed in wet CO₂, whereas it is unvarying in dry CO₂. Comparison between Al burning in CO₂/H₂ and in CO₂/(Ar or He) demonstrates that the oxide cap regression in a wet atmosphere is related to a chemical effect of hydrogen produced in the flame, and then diffusing and reacting at the droplet surface. It is suggested that the adsorption mechanism of H₂ on the Al surface may slow down the contribution of adsorbed oxygen-containing species (CO) to the oxide cap, which would consequently promote its decomposition (removal). Furthermore, the carbon dissolution process is observed in wet and dry CO₂. When the carbon concentration reaches the saturation limit in the burning Al droplet ($x_C = 0.23$ at $T = 2600 \text{ K}$), the excess of carbon is ejected at the surface and forms a solid coating. In the absence of the oxide cap (wet CO₂), the refractory carbon coating prevents strong surface oxidation, and the combustion definitely stops. In the presence of the oxide cap (dry CO₂), the carbon coating reacts and produces an oxycarbide phase which is melted by the laser heating; a new burning regime occurs mainly controlled by direct surface reactions, and leading to the slow oxidation of the droplet and the expulsion of dissolved carbon into CO. Finally, a qualitative model of the combustion of aluminum in CO₂ atmospheres is proposed.

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1. Introduction

Aluminized solid propellants are mainly used in the rocket boosters for heavy-lift launchers such as

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Ariane 5 (MPS-230) or the Space Shuttle (SRB). Because of its high energetic properties, aluminum powder allows a significant increase of the specific impulse of the propellant. For this reason, the combustion of aluminum particles has been strongly studied for these last 40 years [1,2]. However, some phenomena are not yet clearly understood such as the existence of an oxide cap on the Al surface [1–9], or possible fragmentation of the aluminum particle [1,3,4,9–14]. These unsteady processes mainly result from heterogeneous reactions between the gas-phase and the aluminum particle.

The oxide cap evolution is particularly related to the nature of the gaseous atmosphere. The oxide accumulation on the Al surface is generally considered resulting from the retro-diffusion of gaseous aluminum oxides (AlO, Al₂O) from the flame to the particle [15,16]. It was also observed that the presence of nitrogen promotes the oxide cap formation, while the addition of inert gases (Ar, He) seems to inhibit it [3,17,18]. In [2,19], it was shown that the oxide cap size decreases during Al burning in water-containing atmospheres; an oxide cap regression rate was estimated in analogy to the burning rate, and was correlated with the droplet temperature, suggesting a chemical decomposition process of alumina by liquid aluminum into gaseous species.

Apart from the oxide cap, other phases may appear on the Al surface such as the formation of aluminum nitride (AlN) and oxynitride (AlON) during the combustion in N₂-containing atmospheres [2]. The dissolution process of oxygen and carbon inside the particle is also significant. The quantities dissolved in the unburnt residue can reach about 8–10 mol% O, and 18–23 mol% C [18–20]. In fact, the carbon concentration corresponds to the carbon saturation limit in liquid aluminum. It was also found that the excess of dissolved carbon resulting from the permanent consumption of the Al particle is finally ejected at the surface. This carbon coating prevents Al vaporization and stops the gas-phase burning [19]. However, under solid propellant conditions, as aluminum burns in hot gases ($T > 2500$ K), surface reactions may still occur even if there is no more gas-phase combustion. Such a new combustion regime could involve some of the noted unsteady processes (fragmentation).

In [19], we have especially studied the combustion of Al droplets in CO₂/H₂O mixtures because both gases correspond to the main oxidizers produced during the solid propellant decomposition ($x_{\text{H}_2\text{O}} \approx 0.4\text{--}0.5$, $x_{\text{CO}_2} \approx 0.1$). Although high CO₂ concentrations were examined (up to 97% CO₂ = wet CO₂), Al burning in pure CO₂ (dry CO₂) was not studied. Obviously, it could be considered that the combustion in wet or dry CO₂ is the same, but Prentice [4] observed that the presence of small amounts of wa-

ter vapor promote fragmentation in comparison to dry gases. In addition, new propulsion systems using metals burning in pure CO₂ have been recently proposed for Mars mobility or sample return missions [21].

In the present work, the combustion of aluminum droplets in dry and wet CO₂ is investigated in order to clarify the similarities and the differences for both atmospheres. Furthermore, the existence of the second combustion regime suggested by [19] is examined. Accordingly, an experimental setup which has been developed to observe millimeter-sized aluminum droplets burning in cold gases is used to simulate the hot atmospheres under solid propellant conditions by a permanent laser heating. Analyses of the surface phenomena occurring during the combustion and examinations of the unburnt residues are performed and a new qualitative model of the Al burning in CO₂ atmospheres is proposed.

2. Experimental

The experimental setup was detailed previously [2,19]. A 3-mm aluminum droplet is maintained contactless in an aerodynamic levitation system and is heated by a continuous CO₂ laser. Levitation gases are used as the oxidizing environment; for the wet atmosphere, CO₂ is saturated with H₂O vapor by flowing it through a water bath at ambient temperature. For these conditions, wet CO₂ does not contain more than 3 mol% H₂O. The ignition and combustion processes are analyzed both with a high-speed CCD camera (Kodak EktaPro 1000HRCOLOR, 250–1000 frames/s, optical magnification about 1), and with a monochromatic optical pyrometer ($\lambda \approx 0.8$ μm) aiming the bottom hemisphere of the droplet for which we have a clear view of the surface, the oxide smokes being continually ejected in the upper wake of the flame by the levitation gas flow.

As shown in [19] with the same experimental setup, the combustion of aluminum droplets is not self-sustaining for high CO₂ concentrations in H₂O/CO₂ mixtures ($x_{\text{CO}_2} > 0.8$) when the laser is cut off. In the present work, in order to observe the second combustion regime which may begin after the carbon ejection at the droplet surface, the laser keeps on heating all along the burning process with a minimum laser power (at ignition, typically $P = 150\text{--}200$ W; during burning $P_{\text{min}} = 50\text{--}70$ W).

Some unburnt residues are also analyzed by X-ray diffraction. They concern burning Al droplets which have cooled down slowly after the laser is cut off, without fragmentation or violent impact on the levitation nozzle, and which have produced crystallized phases.

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