Spherically-Expanding Flames in Hybrid Aluminum-Methane-Oxidizer Mixtures at Atmospheric Pressure

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1 Introduction

Accidental explosions from gaseous fuels or combustible dusts burning in air continue to be a serious hazard in industry which results in the loss of life and capital [1]. While flames propagating through combustible gas-air and combustible dust-air mixtures have been studied individually, much less work has been devoted to the study of hybrid combustible-dust and combustible gas mixtures. These hybrid mixtures, however, are frequently encountered in industrial settings [2-4].

Hybrid mixtures of metal fuels with hydrocarbon are distinct from hybrid mixtures of organic or plastic dusts burning with hydrocarbon fuels. Plastic and organic dusts compete for the same oxidizers as hydrocarbon fuels, and the addition of solid fuel to fuel-rich mixtures has little effect of the severity of the explosion. Conversely, metals can react with the products of hydrocarbon flames (i.e. steam and carbon dioxide), enhancing the energy density of the reactive mixture.

Researchers have investigated spherically-propagating flames in these hybrid mixtures using constant volume combustion bombs [4]; however, with no optical access to assess the flame symmetry and stability at multiple stages during the propagation, meaningful deductions from the pressure vs time trace are limited. Previous work used a modified Bunsen burner to investigate flames in hybrid aluminum-methane-oxidizer and iron-methane-oxidizer mixtures [5-7]. The results showed at low concentrations of metal fuel, the particles behave as inert additives; however, past a critical concentration, a metal flame front forms and thermally couples to the methane flame. This stabilized configuration, however, may not be fully representative of the entire range of behavior that may occur in transient, freely-propagating flames which are typical in industrial accidents.

The present work aims to address the limitations of both the combustion bomb and stabilized flame experiments by studying flames propagating in latex balloons. The balloons containing the dust-combustible gas mixture provide a nearly isobaric combustion environment, and the use of

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transparent balloons allows for visual analysis of the dust dispersion and subsequent flame propagation.

2.1 Experimental Set Up

In the experiments, the balloon is initially inflated with the desired combustible gas mixture to a diameter of approximately 30 cm in diameter. A high-pressure pulse of the same combustible gas disperses the aluminum powder within the balloon for 0.8 s. There is a 4 s delay after dispersion before the mixture is ignited in order to ensure that the strong turbulence induced by the dispersion has fully decayed. A tungsten wired rapidly heated by the discharge of an electrolytic capacity located at the center of the balloon ignites the mixture. The igniter in these experiments is only strong enough to initiate the gaseous methane flame. This creates the condition for the self-formation of an aluminum flame which has been used in previous hybrid flame experiments [5-7]. The balloon expands as the flame propagates outwards, maintaining nearly isobaric conditions, and bursts as the flame reaches the balloon surface. Detailed schematics of the experimental apparatus can be found in [8].

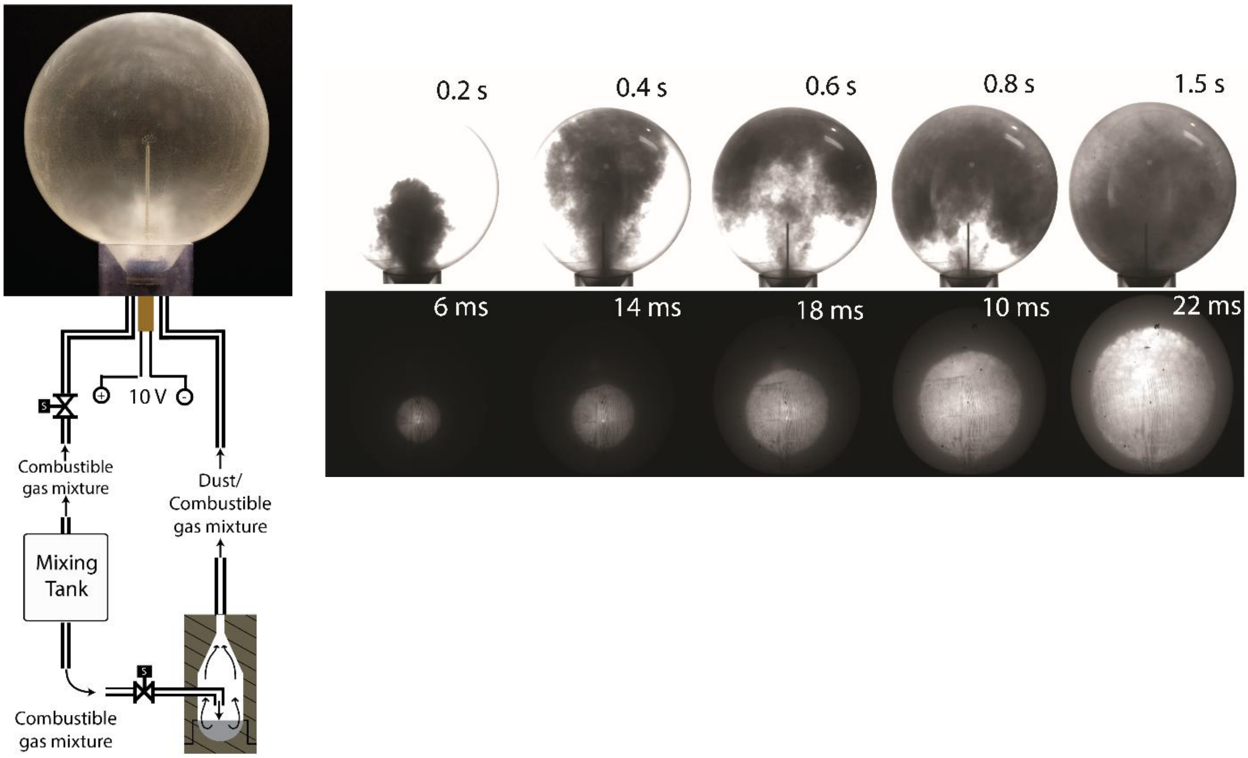

Figure 1: The left part of the figure shows picture-schmeatic of the inflated beloon before dispersion witht the key components shown underneath. The right side of the figure show the dispersion of dust within the balloon (top) and the flame propagation within the balloon (bottom).

2.2 Gas Mixtures and Powders

Three different combustible gas mixtures are studied in the present experiment. The first is a methane-air mixture with an equivalence ratio of 1.1, meaning there is no excess oxygen. The next two mixtures have the same amount of methane, but varying amounts of excess oxygen. The first mixture with excess oxygen is 26% oxygen/74% nitrogen and methane with an equivalence ratio of 0.8, and the second mixture is 30% oxygen/70% nitrogen and methane with an equivalence ratio of 0.7. Therefore, all of the mixtures have nearly the same flame temperature and thermal diffusivity which allows the effect of oxygen concentration to be isolated.

The aluminum powder studied is Ampal 637 (Ampal, NJ) which has a sauter mean diameter of 6.5 microns. This is the same batch of aluminum powder that has been used in a series of previous experiments studying flames in aluminum-oxidizer and hybrid aluminum-methane-oxidizer mixtures. Size distribution and SEM photographs are available in [5,6,8]. As a benchmark, experiments are also conducted with inert spherical alumina powder made by Advanced Materials with a mean particle diameter of 5 microns.

2.3 Measurements Techniques

The concentration of powder in suspension within the balloon is determined using results from a calibration performed within an acrylic sphere having the same size of the balloon [8]. Using a laser-light attenuation probe calibrated for the same aluminum powder [6], a relationship between the mass placed in the powder dispersion unit and the actual concentration of aluminum powder in suspension at the time of ignition was determined. Due to the similar size, it is assumed that the inert alumina powder also followed the same relationship.

The dispersion process and subsequent flame propagation is recorded around 5,000 FPS using a Photron SA-5 video camera. The flame front is tracked by fitting an ellipse to the flame in each frame, and the instantaneous flame radius is taken as the average distance from the flame contour to the center of the ellipse.

The images from high speed videos are also processed for mixtures at low particle concentrations to analyze the flame surface. Particles passing through the flame heat up and emit black body radiation which helps illuminate the flame surface. Therefore, changes in the flame's surface during the propagation can be identified by subtracting subsequent frames in a pseudo-schlieren technique [9]. At higher concentrations, the increased particle density causes more scattering of the light produced from the hot particles, and features on the falme's surface cannot be resolved.

3 Results and Discussion

3.1 The Effect of Oxygen Concentration on Flame Stability

Figure 3a shows typical results of the radius vs time data obtained for three different gas mixtures. As can be seen, the flames in the mixtures with no excess oxygen propagate at a stable speed while the flames in the other mixtures with excess oxygen are accelerating. Sample images analyzing the flame surface are shown in Fig 3b and 3c for mixtures with and without excess oxygen with about 15 g/m^3 of aluminum in suspension. The flames in mixtures without excess oxygen have a smooth surface while the flames in mixtures with excess oxygen have cellular structures on the surface. The flame behavior is the same when reactive aluminum is replaced by inert alumina.

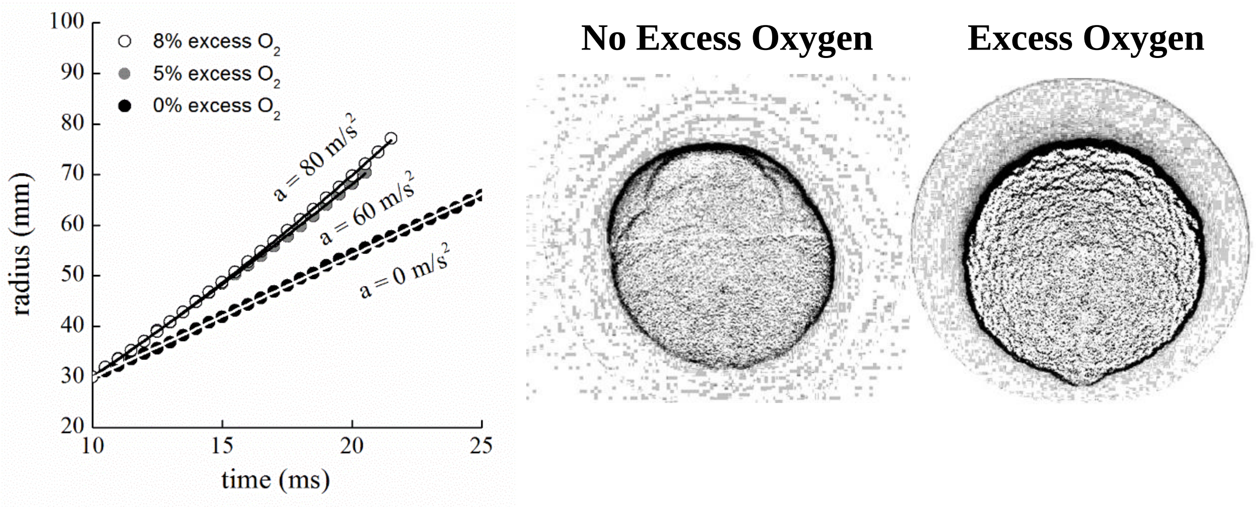

Figure 2: a) Radius-time flame trajectories illustrating both steady (a ≈ 0) and unsteady (a > 0) propagation, b) flame surface in mixtures with no excess oxygen, and c) flame surface in mixtures with excess oxygen.

These affects are attributed the flame becoming hydrodynamically unstable. All spherically-expanding flame become hydrodynamically unstable at a critical radius which results in the spontaneous formation and growth of cells. The increased surface area from the cells increases the reaction rates and causes the flame speed to increase. This claim is further verified by fitting the radius vs time data to: $R = R_\mathrm{c}\left(\frac{S_\mathrm{F} t}{R_\mathrm{c}}\right)^{\alpha}$ for $R > R_\mathrm{c}$, a typical form found in the literature to track the growth of hydrodynamically unstable flames [9]. Here R_c is the critical radius for the onset of instabilities, S_F is the laminar flame speed, and α is the growth coefficient. The results consistently found $R_\mathrm{cl} \approx 20 - 40$ mm, which is consistent with the images in Fig. 2c which shows that flames are cellular during the propagation within the balloon. The results for the growth coefficient, α, show that α remains approximately constant with dust concentration with values consistently between 1.1 and 1.3 for both inert alumina and aluminum. These values for α are consistent values reported in literature for accelerating methane flames at atmospheric pressure [9]. With the source of the acceleration identified, the effect of aluminum combustion on the laminar flame speed, S_F, can be separated from acceleration due to hydrodynamic instabilities.

3.2 The Effect of Powder Concentration on Flame Speed

Figure 3a and 3b show the flame speed as a function of powder concentration for the three different gas mixtures with both inert alumina and aluminum, respectively. The dashed lines represent theoretically calculated flame speeds found using Cantera [10] assuming that the particles in suspension are unreactive. This is done by increasing the heat capacity of the mixture by an amount proportional to the dust concentration. The measured flame speeds for mixtures seeded with inert alumina decrease with increasing particle concentration for all of the gas mixtures, following the dashed lines as expected. Mixtures seeded with aluminum show different behavior depending on whether ether is excess oxygen available.

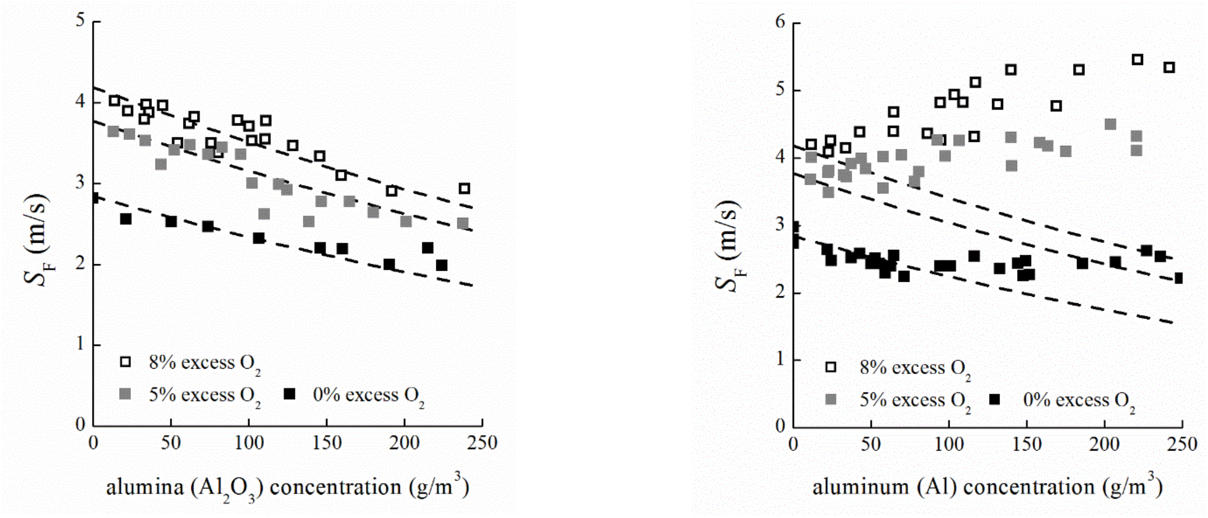

Figure 3: flame speed as a function of a) alumina concentration and b) aluminum concentration

In mixtures without excess oxygen, the flame speed initially decreases with aluminum concentration, following the dashed line which assumes that the particles do not react. These results indicate that the aluminum reaction rates are much slower than the methane reaction rates at these low concentrations. Above a concentration of approximately 100 g/m^3, the flame speed then remains constant for increasing aluminum concentration. This behavior is analogous to behavior previously observed while studying similar hybrid mixtures on a modified Bunsen burner [5,6]. At low concentrations, the aluminum acted as an inert diluent, only contributing by increasing the heat capacity of the mixture. At a critical concentration, an aluminum flame front formed and coupled to the methane flame, forming a double-front structure. Above this concentration, the burning velocity remained constant with additional aluminum in suspension. Although the second flame front cannot be directly observed in the present experiment set-up, the plateau in flame speed past a critical concentration suggests its existence in the present experiments as well.

In mixtures with excess oxygen, the flame speed initially increases with aluminum concentration before also reaching a plateau in flame speed. The plateau in flame speed again indicates that the aluminum and methane flames are coupled at high concentrations, consistent with results from the Bunsen burner experiments [6]. The increase of flame speed for concentrations below the critical concentration needed for coupling, however, was not observed in Bunsen burner experiments for the same excess oxygen mixtures. The different behavior is attributed to the different geometric configurations of the flames. In a Bunsen-type flame, particles reacting behind the methane flame simply flow downstream without influencing the flame. In a spherically-expanding flame, however, particles reacting behind the methane flame are contained within the spherical flame, and any heat release from the burning particles causes the gases to expand and push the methane flame faster.

The difference in behavior between the mixtures with and without excess oxygen at low concentrations is attributed to differences in particle mode of combustion. In mixtures with excess oxygen, the relatively high reaction rates and heat release of aluminum-oxygen reactions compared to aluminum-water vapor/carbon dioxide reactions creates the conditions necessary for particle ignition, defined as the transition from the kinetically-limited combustion regime to the diffusion-limited combustion regime. When particles ignite, they burn as micro-diffusion flames with higher local temperatures which allows particles to react sufficiently fast, even at low concentrations, to influence the methane flame propagation.

The critical concentration behavior observed for mixtures without excess oxygen indicate that the particles are burning in the kinetically-limited regime. In this case, a sufficient concentration of aluminum is needed to raise the bulk temperature of the gas to high enough temperatures to attain sufficiently high aluminum reaction rates.

4 Conclusion

Spherically-expanding flames in hybrid aluminum-methane-oxidizer mixtures are studied using transparent latex balloons. In slightly fuel-rich methane-air mixtures where there is no excess oxygen, the flames propagate at constant speeds. In these mixtures, the aluminum acts as an inert diluent at low concentrations, and only contributes to the flame propagation past a critical concentration of aluminum in suspensions. In fuel-lean, the flames are accelerating during the observed propagation which is attributed to the flames becoming hydrodynamically unstable. In these mixtures with excess oxygen, the flame speed is enhanced even for low concentrations of aluminum in suspensions. The difference in behavior between mixtures with and without excess oxygen is attributed to different particle modes of combustion. The ability of particles to ignite and burn as individual micro-flames when reacting in mixtures with excess oxygen allows them to attain very high reaction rates even at low concentrations. For particles reacting kinetically in mixtures where only water vapor and carbon dioxide are available, a sufficient concentration of aluminum is needed to reach the high temperatures needed to for aluminum to react rapidly.

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