A Study of Stabilized Iron-Methane-Air Flames

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1 Introduction

The combustion of solid fuel-air suspensions occurs in a wide range of applications and has been the subject of many studies. Previous studies have focused on dusts with practical applications, namely energy production (coal, sawdust), mitigation of accidental explosions (sawdust, cornstarch, flour) and the use of dust combustion in propulsion devices (aluminum, boron). Aluminum and boron both produce oxides in the gas phase, while organic dusts (coal, sawdust, cornstarch, flour) burn in a very complex manner, partially volatilizing and burning in the gas phase and partially charring, and burning heterogeneously with surface reaction.

Iron dust is useful for studying purely heterogeneous combustion since all chemical reactions happen at the surface and the oxides produced remain in the condensed phase. Investigating iron dust combustion isolates one phenomenon that occurs in the more complex combustion of organic dusts or boron. The combustion of iron particles has been previously studied in suspensions in air by Sun et al. [1, 2] and in microgravity conditions by Tang et al. [3].

The present study investigates stabilized iron-methane-air flames. The effects of iron concentration on the flame structure, temperature and speed are experimentally observed by optical methods. The results show that iron-methane mixtures can form a double flame front structure separated by a dark zone when the iron concentration is sufficiently high. At low concentrations, iron is believed to slowly oxidize in the hot environment without forming a flame front. Finally, at high concentrations, the flame structure exhibits an instability due to thermo-diffusive effects caused by the increase in the overall heat capacity of the mixture.

2 Experimental Methods

Experiments were performed with a stoichiometric mixture of methane and air. In addition to the methane, the iron fuel concentration varied between 0 and 350 g/m³. The iron powder used was produced by Alfa Aesar, is spherical in shape, with a diameter range of 1-3 micron.

The hybrid Bunsen burner used for this study was previously described in Soo et al. [4]. The iron is delivered to the air knife by a piston, as shown in figure 1, after which the flow is gently expanded and



Figure 1. Schematics of the Bunsen dust burner.

passed through a flow laminarizing tube leading to the nozzle. A remotely controlled propane torch then ignites the combustible mixture.

The concentration of iron is monitored by measuring the attenuation of light, according to the Beer-Lambert law. A laser beam passes through the suspension of dust via a slit in the nozzle just before the flame, where it is attenuated to some degree, and then the beam intensity is recorded with a photodiode detector. Flame images are acquired with a Canon EOS 40D digital camera and the time of the picture is marked for correlating it to the dust concentration. Flame spectra were acquired with an Ocean Optics spectrometer (either a model HR-4000 or model USB-4000) coupled via an optical fiber to a telescope focused on the flame front.

3 Results

Figure 2 shows three flame images acquired at different iron concentrations. Flame A is taken at a concentration of 100 g/m^3 and shows a single Bunsen cone with a significant loading of iron particles emitting blackbody radiation. This image is taken before the formation and coupling of the iron flame, which is depicted by flame B, at about 200 g/m³. In this image, two different flames are visibly separated by a dark zone. The inner Bunsen cone is the methane flame, while the outer cone is the iron flame. Flame D is a close-up image that clearly shows the double front structure. The Bunsen cones are surrounded by a diffusion flame. The co-flow used for this experiment consists of air and is mainly used to contain the products of combustion. The diffusion flame is not of interest in this study. Finally, flame C was taken with an iron dust concentration of about 350 g/m³ and is typical of the unstable cellular structure that develops on the hybrid iron-methane flame surface at higher concentrations. The unstable flame surface is in constant motion, making it impossible to measure a laminar flame speed. The flame quenches when the iron concentration is increased to values above that at which the instability develops.



Figure 2. Flame A is a methane flame loaded with iron particles that do not burn (low concentration). Flame B shows a flame with both an iron and a methane flame front (above the critical concentration). Flame C shows a hybrid methane-iron air flame that exhibits an unstable cellular structure. Flame D is a close-up on the double front structure.

The burning velocity is obtained by dividing the surface area of the cone by the known volumetric gaseous flow rate [5]. Flame images are used to obtain the surface area. The cone profile is manually traced on a touch screen, and this profile is then rotated to obtain the flame surface area. The results are shown in figure 3. The flame speed of a stoichiometric methane-air mixture is about 35 cm/s. The burning velocity of the methane flame first decreases sharply with an increase in iron concentration. The rate of decrease diminishes with increasing iron concentration, particularly with the formation of the double flame front structure at around 180 g/m³. Burning velocities were estimated up to an iron concentration of about 300 g/m³, at which point the instabilities begin to form.



Figure 3. Flame speed as a function of iron mass concentration.

Figure 4 shows a typical spectrum acquired on the flame front using an Ocean Optics USB 4000 spectrometer. The spectrum shows no evidence of atomic lines or molecular bands, with the exception of the sodium Fraunhofer line at 590 nm, arising from a slight contamination of the iron powder. The continuous spectra are fitted to Planck's law of blackbody radiation (assuming that iron particles behave like grey emitters) to obtain the flame temperature. The flame temperatures are determined by fitting Planck's law of blackbody radiation to the spectra produced by the solid particles in the flame

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front. Results are plotted in figure 5 along with the calculated temperatures. Thermodynamic calculations using an equilibrium code give an adiabatic methane flame temperature of 2225 K at stoichiometric conditions. The temperature measured with a small loading of iron particles, below 75 g/m³, is well within the experimental error of the calculated flame temperature. The flame speed decreases when the iron concentration goes above 75 g/m³ and then follows the trend from the thermodynamic calculations.



Figure 4. Spectra of condensed phase particles in the flame front.

Figure 5. Flame temperature as a function of iron mass concentration.

4 Discussion

The experimental evidence presented by Soo et al. [4] on hybrid aluminum-methane-air flames indicated that the combustion of the aluminum particles occurs in a coherent flame front above a critical particle mass concentration. Below that critical concentration, the aluminum particles slowly oxidized in the hot gaseous environment. The transition is clearly correlated with a shift in the temperature data. At the critical mass concentration, there is a sharp increase in the flame temperature, from the methane flame temperature to about 2800 K. Although less obvious, the same interpretation can be extended to the hybrid iron-methane-air flame presented in this work. At a critical concentration, there is enough iron to form an iron flame front that is coupled to the methane flame flame

The flame speed first sharply decreases with an increase in iron concentration and the rate of decrease diminishes at a specific concentration that coincides with the point at which the double flame front appears. The decrease in burning velocity results from a decrease in flame temperature due to the absorption of thermal energy by the solid particles. Increasing the number of particles causes additional heat transfer to the particles, causing the burning velocity to decrease further. The fact that the decrease in burning velocity is less important at higher concentrations, when the double front structure is present, suggests that the iron participates in the flame propagation mechanism by releasing thermal energy.

The flame temperature data show a decrease in temperature with an increase in iron concentration. The double front structure has a flame temperature that is lower than the pure methane flame temperature.

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Although iron participates in the flame propagation mechanism, as observed with the burning velocity, when the double front structure is present, it does not increase the flame temperature. The transition from the slow oxidation of iron to full-fledged combustion must be associated then to the increase in total surface area, which increases the reaction rate sufficiently to form a flame front.

For the case of hybrid aluminum-methane flames, as reported by Soo et al. [4], the flame speed remains constant with an increase in concentration after flame coupling. Thermodynamic calculations show that the flame temperature increases when the aluminum concentration is increased, but in the case of iron, the temperature decreases with an increase in concentration. The reaction of iron with water and carbon dioxide is sufficiently energetic only to diminish the decrease in burning velocity due to the heat absorbed by the greater number of iron particles.



Figure 6. Cellular structure of an iron flame in microgravity [3].

The cellular structure observed at high concentrations is believed to arise from thermo-diffusive instabilities. A similar structure has been observed for pure iron flames in microgravity [3], as shown in figure 6. Thermo-diffusive instabilities typically occur when the flame is deficient in the more mobile reactant, changing the Lewis number of the mixture [6]. Increasing the dust concentration will not vary the amount of gaseous reactants since the methane and air flow rates are kept constant and because dust particles effectively occupy no volume when compared to gases. Thus, an increase in dust concentration doesn't decrease the concentration of oxidizers, the species actually diffusing in the mixture. However an increase in dust concentration will increase the overall heat capacity of the mixture, which in turn affects the heat diffusivity and decreases the Lewis number. When the Lewis number reaches a small enough value, the cellular structure in the flame begins to form.

5 Conclusions

The combustion of iron-methane-air flames falls into three regimes. In the first regime, at low iron concentrations (below 180 g/m³), the flame speed and temperature are strongly dependent on, and inversely related to iron concentration. This is due to the fact that the iron does not take part in the

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combustion and merely acts as a heat sink. In the second regime, at intermediate concentrations (between 180 and 300 g/m³), it is apparent that the iron participates in the combustion process, since the double flame front cannot be attributed to black body radiation. Finally, at high concentrations (above 300 g/m³), the flame becomes unstable. Cells begin to form, and eventually the flame quenches. This cellular structure arises from thermo-diffusive instabilities caused by the increase in heat capacity of the overall mixture at high particle loadings.

The iron mass concentration must reach a critical concentration for the iron particles to start reacting in the hot environment produced by the methane flame. When the iron flame is formed, the heat released by the iron particles participates in the flame propagation mechanism. However, the heat released is not sufficient to fully compensate for the heat absorbed by the extra iron particles. This results in a slower decrease in flame speed with an increase in iron concentration versus the steeper decrease in flame speed when iron is not reacting.

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