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Emission and laser absorption spectroscopy of flat flames in aluminum suspensions

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ABSTRACT

Imaging emission spectroscopy, spatially resolved laser-absorption spectroscopy, and particle image velocimetry (PIV) are applied to a flat flame stabilized in a suspension of micron-sized aluminum. The results from the combination of diagnostics are used to infer the combustion regime of the particles and to estimate the characteristic combustion time of the suspension. It is observed that the reaction zone of the flame in stoichiometric aluminum–air suspensions exhibits strong self-reversal of the atomic aluminum emission lines. These lines also exhibit high optical depths in both emission and absorption spectroscopy. The strong self-reversal and high optical depths indicate high concentrations of aluminum vapor within the reaction zone of the flame at multiple temperatures. These features provide evidence of the formation of vapor-phase micro-diffusion flames around the individual particles in the suspension. In aluminum–methane–air flames, the lack of self-reversal and lower optical depths of the aluminum atomic lines indicate the absence of vapor-phase micro-diffusion flames, and point to a more heterogeneous, and likely kinetically-controlled, particle combustion regime. The reaction zone thickness is estimated from the spatially resolved profiles of aluminum resonance lines in both absorption and emission through the flame. The emission measurements yield a reaction zone thickness on the order of 1.7±0.3 mm in aluminum–air flames, and the absorption measurements yield a thickness on the order of 2.3±0.5. It is demonstrated that the combination of the combustion zone thickness measurement, flame temperatures determined from molecular AlO emission spectra, and particle velocity measurements from the PIV diagnostic permits an estimation of the burning time in the suspension. The burning time in stoichiometric aluminum–air suspensions using the suite of diagnostics is estimated to be on the order of 0.7 ms.

1. Introduction

Micron-sized metal powders, particularly aluminum, are common energetic additives to propellants and explosives. The mass fraction of metal in these energetic compositions can exceed 20%, meaning that the metal burns as a dense suspension in the gaseous products of a hydrocarbon fuel matrix. Understanding the combustion behavior of metal particles in a dense suspension is central to the goal of predicting and tailoring the performance of metalized explosives and propellants. Metals are also increasingly being investigated for use as recyclable fuels [1–3], which would be burned in dense suspensions to achieve high power densities and ensure flame stability.

The combustion physics of a dense suspension of micron-sized particles can differ considerably from the combustion of large, individual, isolated particles, which have been the dominant subject of experimental work in metal combustion [4]. While large particles burn primarily in a diffusion-controlled regime of combustion, combustion of micron- and submicron-sized particles can be limited by heterogeneous kinetics or they can burn in a transitional diffusion-to-kinetic regime [5]. The combustion characteristics measured from single particle studies, such as combustion time and combustion regime, are often extrapolated to particles in the dense suspensions encountered in the majority of practical fuel systems. However, these characteristics are also functions of ambient temperature and oxidizer concentration and, therefore, will depend on the fuel particle concentration [6,7]. The extrapolation of characteristic combustion data from single-particle studies to dense metal suspensions, where temperatures can exceed 3000 K, will, in general, result in erroneous predictions. Thus, it

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is imperative to have experimental techniques to determine combustion characteristics of bulk reacting suspensions.

In our previous work, stabilized Bunsen-type flames were used to study combustion characteristics of aluminum and iron suspensions in oxygen and in the products of hydrocarbon flames [8–11]. Other studies have used freely-propagating flames in tubes [12] or, more recently, transparent latex balloons [13], to study flame propagation speeds, laminar burning velocities, and the formation of instabilities. The burning velocities measured in these studies permit a more realistic estimation of the characteristic combustion time of the suspension compared to experiments with individual particles.

However, the determination of key combustion characteristics in suspensions of metal particles is difficult due to the inability to isolate and observe individual particles in the cloud. Optical diagnostic techniques to extract these parameters must be tailored to the specific nature of metal suspension combustion which can be optically dense, contain multiple temperatures, have intense luminosity, and cause strong multiple scattering. These phenomena are not often encountered in hydrocarbon combustion systems and largely prevent the use of “off-the-shelf” approaches to the study of combustion in suspensions.

Tailoring standard diagnostic techniques to metalized suspensions is often not straight forward. For example, recent studies show that coherent anti-Stokes Raman scattering (CARS) for gas temperature measurements in heavily metalized propellant flames may only be feasible with the use of femtosecond and picosecond laser pulses [14]. Using a technique like planar laser induced florescence (PLIF) to study the combustion of individual burning aluminum particles [15] is not realistic for quantitative measurements in suspensions due to the high particle number density. However, PLIF may still be useful as an imaging technique in these multiphase environments [16].

Emission spectroscopy was previously applied to aluminum Bunsen flames to compare temperatures from gaseous AlO band spectra and condensed-phase continuum emitters, in an attempt to diagnose the particle combustion regime in the suspension and to compare temperatures to equilibrium calculations [17]. Line-of-sight integrated emission measurements from the cylindrically symmetric Bunsen flame cones required the use of a reverse Abel transform to reconstruct the radial profiles of spectral emission intensity to determine the thermal structure of the flame and the width of the combustion zone. The high optical thickness of both the condensed phase spectra and the AlO molecular bands, however, rendered the Abel reconstruction performed in [17] unreliable. The experiment was further complicated by the presence of strong multiple scattering of light by aluminum and aluminum oxide particles, which contaminated local emission spectra with the radiation from different regions of the flame. The accuracy of temperature measurements using continuum and molecular AlO spectra was also found to be insufficient to resolve the temperature difference between diffusively burning particles and the bulk flame temperature in the fuel-rich flames [17]. Due to these difficulties, the study was not able to draw definite conclusions about the regime of particle combustion in the suspension.

In the present study, a counterflow dust burner is used to create a flat aluminum dust flame with a one dimensional flame geometry that eliminates the need for spatial reconstruction using the Abel transform. Using the flat flame, an emission diagnostic technique that permits the direct qualitative determination of the regime of aluminum particle combustion in dense suspensions is demonstrated. The technique is based on the spectroscopic detection of self-reversal of the atomic aluminum emission lines that occurs due to inhomogeneous metal vapor temperatures near the micro-diffusion flames that surround the aluminum particles and in the bulk gas of the suspension. The diagnostic technique is also used to measure the combustion zone thickness. The length of the region where the lines are self-reversed serves as a measurement of the reaction zone and largely avoids the difficulties of interpreting the emission spectra in the presence of multiple scattering.

These measurements are compared to those obtained using a complementary diagnostic technique of spatially resolved absorption spectroscopy to detect the aluminum resonant lines. Using a broadband laser as a spectral source, the effects of multiple scattering are greatly reduced, and the length of the region where aluminum is detected serves as a measurement of the reaction zone. Combined with a particle image velocimetry (PIV) technique for estimating the particle residence time in the combustion zone and temperatures measured from AlO molecular spectra, the suite of diagnostics permits the characteristic combustion time of a dense aluminum suspension to be estimated.

2. Experimental methods

2.1. Counterflow burner and aluminum/gas mixtures

The spectroscopic study is performed on flat aluminum dust flames stabilized using a counterflow dust burner (see Fig. 1(a)). The design of the apparatus is described in [9], where it was used to measure burning velocities in aluminum clouds using a PIV technique. For this study, the apparatus is modified to increase the flame planarity. The coaxial flow nozzle on the opposing jet in [9] is removed in order to eliminate recirculation of the combustion products on the flame periphery because it interferes with concentration and spectral measurements. The air in the coaxial flow around the bottom nozzle is also replaced with inert nitrogen to prevent formation of the diffusion flame often enveloping flat flames at fuel-rich conditions. Concentration of the particles is monitored by a laser attenuation probe described in previous work [18]. In the present study, the laser beam probe passes just above the exit of the bottom nozzle as shown in Fig. 1(b). An attempt was made to keep the concentration of aluminum in the suspension stable at around 300 g/m^3 for all measurements. Considering the accuracy of the dust concentration measurements, the actual concentration varies between 270 and 330 g/m^3.

The combustion in the aluminum suspension is studied in two gaseous environments: air and a stoichiometric methane-air mixture. The gas flow rates are monitored using factory calibrated electronic flow meters and rotameters. The oxygen concentration in aluminum–methane–air mixtures is monitored by an in-situ oxygen analyzer (Oxigraf) to determine the equivalence ratio.

2.2. Aluminum powder

Unlike our previous studies of stationary aluminum dust flames that used Ampal 637 aluminum (Ampal, Inc.) with nodular shaped particles with a size around 6 μm [8,9,11], the present study employs H-2 aluminum powder produced by Valimet Inc. (Stockton, CA) with spherical particles and a narrow particle size distribution. The particle size distribution in the H-2 powder from laser diffraction measurements is provided by Valimet and shown in Fig. 2. The arithmetic mean diameter (d_{10}) in the H-2 aluminum powder is reported to be around 4.2 μm.

2.3. Emission imaging spectroscopy setup

The general schematic of the imaging spectroscopy setup is shown in Fig. 1(c). The setup consists of a 250 mm focal length spherical lens that images the flame onto the entrance slit of an imaging spectrometer with a focal length of 0.3 m equipped with a CCD array. Due to the internally recessed focal plane of the instrument, the CCD array detector is coupled to the spectrometer by a de-magnifying image relay consisting of two imaging