



# Laminar flame speeds of nano-aluminum/methane hybrid mixtures



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## ABSTRACT

An existing flame speed bomb was utilized to study the fundamental phenomena of flame propagation through a uniformly dispersed aerosol with a relatively small mass of metal nanoparticles. The aerosol was dispersed with a burst of gas and then allowed to settle for at least 45 s, to ensure that the conditions inside the test chamber were quiescent and uniform. Extinction of the light from a HeNe laser was used so that the mass of suspended nano-particles with a fundamental size of 100 nm could be determined as a function of time prior to combustion, and the suspended mass of aluminum (up to 90 mg) was measured in situ during an experiment. A particle size distribution was measured as well, resulting in an average pre-combustion agglomeration size of 446.1 nm. Two series of experiments were performed with CH<sub>4</sub> fuel, both at stoichiometric conditions: one with the mixture in air and the second with the mixture in a 70/30 N<sub>2</sub>/O<sub>2</sub> mix. The results herein show a maximum decrease in flame speed, 5–7% for the baseline mixture, when nano-aluminum was introduced at the suspended masses utilized in the present study. For the 70/30 N<sub>2</sub>/O<sub>2</sub> mixture, the aluminum resulted in a maximum decrease of 5 cm/s from the baseline value of 80.5 cm/s; and in the air mixture, a 2 cm/s maximum decrease from 35.3 cm/s was observed. Simple modeling of the process extremes indicates that the observed decrease in flame speed is somewhere between the limits of simple particle heating with no particle reaction and the kinetics limit where the Al reacts in the gas phase. Interestingly, the trends qualitatively matched those of the kinetics model applied herein. It was also found that the addition of nanoparticles caused the flame to become unstable much sooner when compared to the baseline mixture of CH<sub>4</sub>/air.

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## 1. Introduction

Fundamental research on particle combustion, with a parent fuel or without, is of great scientific interest because such particles are increasingly being investigated as additives in applications such as solid and liquid rocket propellants, among others. Additionally, having fundamental information of how particles combust may provide the ability for more accurate models in the process safety field. The near-term objective of this study was to establish a method of measuring laminar flame speed – synonymous with burning velocity, flame velocity, and normal combustion velocity – of an aerosol mixture to a higher degree of accuracy than has been achieved in previous studies [1]. For the purposes of this study, laminar flame speed is defined as “the velocity, relative to the unburnt gas, with which a plane, one-dimensional flame front travels along the normal to its surface” [2]. Accomplishing this objective will provide a means to gain a better understanding of the

mechanisms that drive combustion in the presence of solid particulates and, in general, heterogeneous combustion in aerosol and hybrid particle-fuel systems for a number of applications. It is suspected that in most literature sources the largest amount of experimental error comes from the injection method and the resulting lack of knowledge about the particle size distribution and concentration of particles in the mixture at the time of reaction. Additionally, research on heterogeneous mixtures in constant-volume bombs is mostly limited to pressure data with an optical access (if any) just large enough to verify ignition has occurred. Due to a lack of optical access, there is a corresponding lack of information about the level of turbulence at the time of ignition, as well as the homogeneity of the aerosol. This uncertainty is exacerbated by the common method of introducing the dust and igniting it in these experiments, which is to force it in with a blast of compressed air and to ignite it within seconds or even milliseconds, at the most. However, this study is not intended to be a comprehensive literature review; therefore, for more information on the experimental facilities and procedure of standardized explosion testing see [3–5], and additional information about aluminum-specific explosion testing can be found in other references [6–9]. Although the

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present study is more concerned with trace levels of solid particulates, there are nonetheless some similarities with conventional dust-explosion studies in constant-volume vessels.

In conventional dust explosion studies, the dust is introduced into a combustion vessel via a blast of compressed air and then almost immediately ignited, for example after 60 ms in Dahoe et al. [3] and 400 ms in Cashdollar [4]. Because the aerosol is ignited so quickly, it is known that the atmosphere inside the combustion vessel is not quiescent and that the dust may not be equally distributed. Some studies have attempted to measure the turbulence inside of their vessels to correct their burning velocities to a maximum effective burning velocity, that is thought to be related to turbulent burning velocity; however, even if turbulent burning velocity could be determined, there is currently no widely accepted method of extracting laminar flame speed from it [10,11]. This common method has proven adequate if the objective is to compare dusts on a relative basis; however, understanding of fundamental combustion mechanisms, particularly with lower loadings of particulates, requires finer control over the experiment. There have been attempts to study laminar flame speed of hybrid mixtures using the Bunsen burner method, for example Dahoe et al. [12] and Soo et al. [13], which has its own set of limitations such as non-ideal curvature, but no study has attempted to measure laminar flame speed in an optical, spherically expanding flame. Optical access is extremely important to determine if the flame is properly spherical and if there are any instabilities because if either of these conditions are not met, then neither the pressure method nor the optical method of determining laminar flame speed is valid [14].

To this end, the authors have established a test method for aerosol mixtures that tries to have a high level of control on the mixture uniformity, a knowledge of the particle concentration at the time of ignition, a quiescent environment, and ample optical access for various optical techniques. In the present study, modifications were made on an existing gas-phase, spherical flame vessel to introduce dust into a controlled environment in a repeatable fashion to measure the flame speed using existing optical methods.

The present paper is set up as follows: the method to inject dust, characterization of the nano-aluminum, extinction measurements, and the aerosol flame speed system are discussed first. The experimental extinction, flame speed, process-limit modeling, and flame structure are then presented and discussed. Finally, the results are concluded with a few remarks.

## 2. Experimental facility and approach

The laminar flame facility used in the present study is a 28.1-L, 35.6-cm long cylindrical vessel originally designed to be filled with purely gas-phase components up to a maximum initial pressure of 5 atm. The vessel is filled using the partial pressure method using two pressure transducers with precisions of 0.013 kPa and 0.689 kPa, the low-pressure and high-pressure transducers, respectively. Fuel–air mixtures are ignited using a central ignition system, and the resulting flame propagation is captured using a high-speed camera in a Z-type Schlieren setup. For additional information regarding the existing flame speed facility, including equipment, testing procedure and theory, see the earlier works of de Vries [15] and Lowry et al. [16]. Provided below are details on how the facility was modified to allow for the controlled introduction of suspended solid particles.

### 2.1. Dust injection

This study is based on a variant of a “direct-injection” method [17]. The objective during the system design was to create a method of performing tests relatively quickly, while still maintaining a highly repeatable experiment from test-to-test. One way of

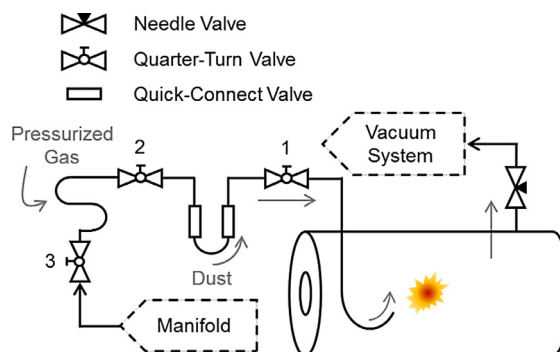


Fig. 1. Piping schematic for gas filling and aerosol dispersion.

accomplishing the objective was to minimize the amount of piping that the dust would have to flow through to get to the experimental vessel. The schematic for this design is shown in Fig. 1.

The dust is loaded into the system by removing the U-Pipe (Fig. 1), weighing the empty U-Pipe, and scooping dust into the U-Pipe until the dust mass is at a predetermined amount. The weighed mass of dust in the U-Pipe, herein referred to as the loading, is only used as a general guide to how much suspended mass will be put into the vessel. There is a positive correlation between the loading and the final suspended mass; however, the chosen method of measuring the suspended mass within the system, using optical extinction as discussed later, removes the need to stringently develop a relation between the loading and the suspended mass because the optical measurement is performed independently from the loading.

To inject the dust loading, the piping between valves 2 and 3 (Fig. 1) is highly pressurized, and the experiment begins when valve 2 is opened, allowing the high-pressure gas to induct the particles into the combustion vessel. The system is purged repeatedly to remove as much excess dust as possible in between tests. This procedure has been shown to result in suitably repeatable experiments using extinction tests which are described below.

### 2.2. Nano-aluminum characterization

For all experiments herein, the chosen dust was a 100-nm nano-aluminum type purchased from US Research Nanomaterials, Inc. A sub-micron dust was used because of the nature of the testing procedure, which can occur over several minutes. Initially, nano-sized particles were preferred primarily because of their aerodynamic/drag advantages, but later experiments demonstrated their advantages with regard to possibly burning quickly, as discussed later. To elaborate, the particles must be very small so that they stay suspended long enough for the air to become quiescent prior to flame initiation, and so that the particle surface area is maximized to promote a quick reaction. To verify that the fundamental particle size of the nano-aluminum is close to the manufacturer's claim, the material has been analyzed using a transmission electron microscope (TEM) in Fig. 2. To isolate the agglomerate in panel (a) of Fig. 2, the nano-aluminum was first suspended in nitromethane and sonicated to break up any large agglomerates. This mixture was then put into a carbon mesh and the nitromethane evaporated off. The mesh containing the nano-aluminum was then placed into the TEM for imaging [18]. The TEM image of the agglomeration is not representative of those seen in this study. That agglomeration size of the suspended particles is investigated below. This TEM image is useful to get an estimate of the fundamental particle size.

Based on these images, the fundamental particle size is close to the manufacturer's claim, and it can be assumed that there is

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