



## Full Length Article

## Effects of pressure and preheating temperature on the laminar flame speed of methane/air and acetone/air mixtures

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## ABSTRACT

Laminar flame speeds of acetone/air mixtures have been investigated at elevated temperature and high pressure conditions in regards to the importance of this intermediate oxygenated molecule in hydrocarbon oxidation. Experiments were performed in an elevated pressure combustion vessel specifically designed around a concept of a premixed Bunsen flame. In a first step, measurements of the laminar flame speed of gaseous CH<sub>4</sub>/air mixtures were performed with OH<sup>•</sup> chemiluminescence and OH-PLIF methodologies and compared with literature data in order to validate the experimental setup. The effects of the preheating temperature (373–523 K), pressure (0.1–1.0 MPa) and equivalence ratio (0.6–1.3) on the laminar flame speed of acetone/air mixtures were then examined. The experiments were finally complemented and compared with numerical simulations conducted with Cosilab commercial software package using published detailed chemical kinetic mechanisms for proposing a correlation relationship of the acetone/air laminar flame speed with pressure, temperature and equivalence ratio.

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## 1. Introduction

With the increasingly stringent regulation of worldwide gas emissions and the need for fuels diversification, it is expected that the use of renewable fuels in the aeronautic and automotive transportation will increase. In recent years, biomass derived fuels (bio-fuels) have gained much attention as potential alternatives to petroleum based fuels. Apart from the advantage of renewability, biofuels have shown to be sustainable and less harmful to the environment; especially those derived from 2nd generation biofuels where sustainable lignocellulosic biomass is used as feedstock. One of the common features of biofuels is that they are all oxygenated hydrocarbons, containing oxygen as an additional element in their molecular composition. This feature distinguishes them from the hydrocarbons in conventional petroleum based fuels whose combustion chemistry has for a long time been studied. On the one hand that the use of oxygenated hydrocarbons in combustion offers significant potential for reduction in particulates and NO<sub>x</sub> emission as compared to conventional hydrocarbon fuels. On the other hand, the incomplete combustion of oxygenated hydro-

carbons may contribute to the emission of small amount of oxygenated hydrocarbons themselves or their intermediates or even harmful chemical components for environmental safety and human health. Therefore, it is crucial at the first step to understand the fundamental combustion properties of different classes of oxygenated molecules playing a key role on their formation and consumption in flames.

Laminar flame speed is one of the fundamental characteristics of the fuel/air mixtures combustion phenomena related to reaction mechanism, diffusion probability and heat release rate etc. [1–3]. It is also of practical significance in designing advanced combustion devices running at high efficiency and low NO<sub>x</sub> emission without flashback and blow-off risks [1,2]. The present study aims to address several key issues related to the laminar flame speed of acetone which is considered as a C<sub>3</sub> oxygenated hydrocarbon (i.e. representing the smallest hydrocarbon regrouping alcohol isomers, aldehyde and acetone structures). This molecule has been selected as a good candidate to build a first combustion mechanism block required for the development of more accurate reaction models for larger oxygenated hydrocarbons. Acetone is also a convenient fluorescence tracer and has been extensively used in acetone PLIF technique for flow visualization and combustion study due to its photophysical advantages [4]. As a result, the chemical behavior of acetone/air mixtures as tracer on the target fuel in hot oxidizing environment must be identified to minimize its perturbation on

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the target mixture and flames. Experimental measurements of laminar flame speed of acetone with high fidelity are therefore needed in operating conditions relevant to industrial burners.

An analysis of the published literature reveals that the laminar flame speed of acetone/air mixtures has been already visited by a few authors [4–10] in recent years. For instance, the fundamental physical parameters such as ignition delays, species measurements and laminar flame speed in acetone/air flames were deduced from studies performed by Pichon et al. [8]. Chong and Hochgreb [4] also performed laminar flame speed measurements using the particle imaging velocimetry (PIV) optical technique in a jet-wall stagnation flame at atmospheric pressure and for an inlet temperature of 300 K. In the most recent study by Nilsson et al. [7], acetone/air laminar flame speeds were measured using a perforate plate burner at atmospheric pressure, and the initial fuel/air mixture temperature range was extended for the first time to higher values from 298 to 358 K. However, large discrepancies between laminar flame speeds were observed from these experimental investigations. By comparing previous experimental results of acetone/air mixture at 298 K and atmospheric pressure conditions it was found that the gap between previous measurements could be up to 40% [4–8]. Furthermore, few experimental investigations at larger pressure and preheating temperatures >400 K are displayed in literature. As a consequence, for a better understanding of the acetone combustion process, it is necessary to complete these acetone/air flame studies over much wider temperature, pressure and equivalence ratio ranges.

By definition, the laminar flame speed,  $S_u$ , is the velocity relative to the unburned gas in which a planar, one-dimensional flame front travels along the normal to its surface [11]. The main experimental methods for measuring laminar flame speeds involve stretched (counter flow or stagnation) [12–15], spherical time-varying (spherical bomb) [16–21], curved (Bunsen type) flame [1–2,22–28] and heat flux method [3,7,29–30]. Each of these approaches has its own advantages and range of applicability. For instance, the advantage of spherical bomb method is the capability to perform high pressure measurements; meanwhile one of its main drawbacks is the difficulty in measuring flame speed at relatively high reactive temperature. At high temperature, the auto-ignition time of the reactants is reduced and can be comparable to the residence time required to ensure no motion of the reactants in the combustion chamber. The counter flow method needs extrapolation to zero stretch and this technique is not adapted for reactive mixtures featuring high burning velocity. The heat flux method are considered as a promising method as the flame is quasi-adiabatic and flat, thus a direct determination of the laminar flame speed is possible without correction for stretch. However, the heat flux method has a limitation in measured velocity, so that  $S_u$  up to 40–70 cm.s<sup>-1</sup> can be measured, and above that, the flame area becomes disturbed by the presence of the perforated burner plated. Premixed Bunsen flame has been one of the pioneer investigated flames in the past. The Bunsen flame is controlled by rather complicated physical phenomena and the main disadvantages include modifications due to heat losses at the rim of the nozzle, variable stretch and curvature effects on the flame tip and potential modifications of the equivalence ratio of the fuel/air mixture due to fresh air entrainment around the flame [27,31]. Nevertheless, the same as the other measurements methods Bunsen flame experimental configuration are still be used recently as it offers attractive properties to measure laminar flame speeds in regards to its simplicity to study fuel/air mixtures with variable preheated temperature and elevated pressure conditions. Furthermore, the combination of this type of burner and an additional controlled evaporator and mixer system may help to achieve vaporized large molecule weight liquid fuels/air mixtures. In the recent work by Wang et al. [2], comparisons between OH-PLIF Bunsen flame

method and heat flux method were performed by measuring laminar flame speed of syngas/air mixtures at the same measurements conditions. It is found that the flame speeds measured by attentively employed Bunsen flame method can be closed to the adiabatic laminar burning velocity measured by heat flux method. More recently, by using Bunsen flame method Gao et al. [28] investigated the effect of ozone addition on laminar flame speed of syngas/air mixtures in low and moderate pressure conditions. Sun et al. [32] measured flame speed of CO/H<sub>2</sub>/O<sub>2</sub>/H<sub>2</sub> mixtures using Bunsen flame method to validate new developed chemical mechanisms. A summary of available experimental investigations in the last ten years on laminar flame speed measurements using Bunsen flame is given in Table 1. According to the literature, simple gaseous fuels are mostly often used to perform laminar flame speed measurements in Bunsen flames at atmospheric pressure. Unfortunately, applications of Bunsen burners for studying complex fuels such as vaporized liquid fuels at elevated pressure and preheating temperature conditions are still limited. In the current work, the Bunsen flame concept is optimized to perform laminar flame speed measurements of vaporized acetone fuel at various preheating temperatures in elevated pressure environments (up to 1.0 MPa).

The structure of this paper is described as follows. First of all, a new high-pressure laminar flame burner has been designed for monitoring laminar flames of gaseous or liquid fuels over a wide range of operating conditions including preheating temperature, pressure and equivalence ratio. The high pressure combustion chamber, fuel vaporization system and optical diagnostic systems are presented in detail in Section 2. The experimental methodology used for the measurement of the laminar flame speed is then described in Section 3. The error sources associated to these techniques are discussed and a new image processing method is proposed. The numerical results are presented and discussed in Section 4. Measurements of the laminar flame speed at high pressure for primary reference CH<sub>4</sub> fuel are firstly presented to validate the high-pressure Bunsen burner and to complement experimental data published in the literature. Then the experimental work is extended to the measurements of the laminar flame speed of acetone/air mixtures at pressure ranging between 0.1 and 1.0 MPa, temperature of 373–550 K and equivalence ratio of 0.6 to 1.3. A comparison between these measurements and numerical simulation using a detailed chemical mechanism is finally accomplished. The last section is devoted to provide improved empirical correlation for laminar acetone/air flames, as well as to study their temperature and pressure dependencies. For this purpose, an analysis of the effects of pressure, temperature and equivalence ratio on the laminar flame speed and an empirical flame speed correlation in function of these parameters are proposed.

## 2. Experimental details

### 2.1. High-pressure Bunsen burner

The burner assembly and pressure chamber used to produce the high-pressure combustion are illustrated in Fig. 1. The chamber is designed to burn up to 3.0 MPa at a maximum wall temperature of 600 K. An axisymmetric premixed burner is designed and developed to generate a steady conical laminar premixed flame stabilized on the outlet of a contoured nozzle in a high pressure chamber (Fig. 1). The shape of the axisymmetric central contracting nozzle is designed with a fifth-order polynomial to reduce the boundary layer thicknesses by accelerating the flow and providing a flat velocity profile at the nozzle outlet. The contoured nozzle has an outlet diameter of  $d_1 = 10$  mm and a contraction ratio of  $\delta = (D/d_1)^2 = 49$ . A second concentric contoured nozzle of outlet diameter of  $d_2 = 10.6$  mm surrounding the central nozzle

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