

Experimental investigation of the laminar burning velocities of methanol, ethanol, *n*-propanol, and *n*-butanol at high pressure



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HIGHLIGHTS

- Methanol, ethanol, *n*-propanol, and *n*-butanol flame speed experiments were conducted.
- Comparison of flame speeds for potential bio-fuel candidates.
- Comparison of results from this study and published literature.
- Simulations tend to underpredict measured values at high pressure.
- Sensitivity analysis suggests further investigation of the pressure dependence.

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ABSTRACT

Due to their increasing share, the combustion of alternative fuels and in particular oxygenated, bio-derived fuel components need to be characterised. The laminar burning velocity is one key parameter for the characterisation of fuels, and it also serves as an important quantity to validate chemical kinetic models. Methanol, ethanol, *n*-propanol, and *n*-butanol laminar burning velocities experiments were conducted in a spherical combustion vessel at an unburnt temperature of 373 K and a pressure of 10 bar. Measured burning velocities from this study and from the published literature are compared with numerical simulation data from published chemical mechanisms. The models tend to underpredict the experimentally measured values. A sensitivity analysis suggests further investigation of the pressure dependence for the fuel specific reactions with hydrogen and hydroxyl radicals.

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1. Introduction

The laminar burning velocity is one key parameter for the numerical simulation of gasoline engine combustion processes. Dependent only on the mixture composition, temperature, and pressure, it serves as a fundamental property of a fuel. This makes the laminar burning velocity an important global kinetic parameter for assessing fuel reactivity and for validating chemical kinetic mechanisms. Alcohols, being oxygenated fuel components, can be used as octane booster or as a biofuel component for fulfilling legislative regulations. Production of alcohols can be realised via the fermentation of sugars, starches, and lignocelluloses or for example via methanol synthesis with synthesis gas obtained from agricultural feedstock. Blending methanol into gasoline as an octane improver has been well established in the past. In addition, ethanol is frequently used as a blending component for blend rates up to

10%vol_{liq}. *n*-Propanol and *n*-butanol are interesting alternative fuel components as a supplement to methanol and ethanol in spark ignition engines.

The evolution of laminar burning velocity for alcohol components has been investigated mainly at atmospheric pressure in the past, and as a consequence experimental data for high pressure are sparse. Here, the laminar burning velocities and Markstein lengths of different alcohols are investigated by conducting experiments under high initial pressure of 10 bar and an elevated temperature of 373 K. The experimental setup consists of a spherical closed pressurised combustion vessel with optical access. Tested fuels in the presented work are methanol, ethanol, *n*-propanol, and *n*-butanol. Several kinetic models from the literature are compared to the new experimental data. In addition, the chemical mechanisms are compared to premixed counterflow burner data for laminar burning velocities under ambient pressure and elevated temperature of 343 K by Veloo and Egolfopoulos [1] for *n*-propanol and by Veloo et al. [2] for methanol, ethanol, and *n*-butanol flames.

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2. Experimental setup and conditions

Experiments were performed using the closed-vessel method in order to acquire experimental data under elevated pressures and temperatures. The experimental setup is shown schematically in Fig. 1. The principle of the setup has been described in detail by Hyvönen [3]. The internal shape of the pressure vessel is spherical with an inner diameter of 100 mm; quartz windows with a diameter of 50 mm are positioned on opposite sides. The outward location of the flame is imaged using a Schlieren cinematography system combined with a high-speed CCD camera (Photron Fastcam 1024PCI). The camera maps a $50 \times 50 \text{ mm}^2$ area with a resolution of 384×384 pixels and a frame rate of 5000 frames per second (fps). The Schlieren system consists of a HeNe laser with a wavelength of 632.8 nm as a light source, a microscope objective, a polariser to adjust the intensity, spherical lenses, a pinhole with a diameter of 0.6 mm, and an optical filter for preventing overexposure of the camera from flame radiation.

An external mixing vessel, directly connected to the combustion chamber via pipes, is used for an external preparation of the air/fuel mixture. This is a major adjustment in comparison to the experimental setup introduced in prior studies, where a fuel storage tank was directly mounted to the combustion vessel [4]. In the present setup, the fuel storage tank is joined to the mixing vessel through a fine needle valve. The fuel storage, the external mixing vessel, and all pipes in contact with fuel are heated to prevent fuel condensation. The required amount of fuel can be calculated as a function of equivalence ratio, temperature, and pressure. Thus, the partial pressure method can be used for accurately measuring and controlling the filling process. Commercially available technical air, used as oxidiser, is slowly filled into the mixing vessel at isothermal conditions until the final pressure is reached. The amount of fuel/air mixture prepared inside the external mixing vessel lasts for several consecutive runs in the combustion vessel. This guarantees a well defined mixture for each equivalence ratio.

The modified setup significantly improves the quality of the experimental results by further reducing standard deviations. Compared to data presented by Jerzembeck et al. [5], who used a syringe system, a reduction in relative standard deviations was achieved from around 10% to less than 1% for stoichiometric mixtures at comparable conditions.

Before sparking, the heaters are turned off and the mixture is allowed to settle. A two-step ignition system is used for igniting the mixture at the centre of the vessel with extended spark plug electrodes of 1 mm diameter.

The experimental setup was verified against laminar burning velocities presented in the literature for methane/air mixtures at an initial pressure of 1 atm and an initial temperature of $\sim 300 \text{ K}$ (298–300 K). Fig. 2 shows a compilation of experimental data from several research groups, which were also acquired with the

combustion vessel method [6–11]. Data acquired with the counter flow setup, presented by Dong et al. [12], is also plotted. This setup is the same as the one used for the alcohol/air measurements by Veloo and Egolfopoulos [1] as well as by Veloo et al. [2]. New methane/air flame data acquired with the present combustion vessel setup are included for verification. Experiments were performed with technical air, which has an oxygen mole fraction of $[\text{O}_2]/([\text{O}_2] + [\text{N}_2]) = 0.205$. In order to simplify the comparison between the new data and the literature data, the new data were scaled to real air conditions with an oxygen content for real air of $X_{\text{O}_2} = 20.94\%$. Results from numerical flame simulations with the GRI 3.0 chemical kinetics mechanism [13] for methane/air mixtures, both for real air as well as technical air conditions, were used for the scaling. For completeness, the original data and the scaled data are shown in Fig. 2. The scaled data points from the present work agree very well with combustion vessel results from Rozenchan et al. [9], Hassan et al. [10], Chen [8], and Law [11]. The scaled data points also agree very well with the counter flow results from Dong et al. [12]. At an equivalence ratio of 0.7, the results are in a range between 13.4 cm/s ([10]) and 16.2 cm/s ([11]). At stoichiometric condition, the range is even smaller between 34.6 cm/s ([11]) and 35.8 cm/s ([9]). At a rich fuel/air mixture of 1.3, the values are between 23.8 cm/s ([11]) and 26.2 cm/s ([9]). For equivalence ratios from lean to stoichiometric, the laminar burning velocity results from Gu et al. [6] as well as from Bradley et al. [7] are significantly faster than the others. For richer conditions the overall scatter increases for the different sets of data.

The basic physical properties and octane numbers of methanol (CH_3OH), ethanol ($\text{C}_2\text{H}_5\text{OH}$), *n*-propanol ($\text{C}_3\text{H}_7\text{OH}$), and *n*-butanol ($\text{C}_4\text{H}_9\text{OH}$) are listed in Table 1. The production uncertainty of the mole fraction of oxygen in the technical air used here in all experiments as specified by the manufacturer has maximum deviation of 5%. Tests with a gas chromatography system (Agilent 6890N GC) for various batches of technical air have shown that uncertainties are less than 2.0%.

The experimental conditions of the fuel/air mixture inside the combustion vessel prior to ignition were set to a temperature of 373 K and a pressure of 10 bar. The temperature was controlled with thermocouples of type K and the pressure was monitored with Kistler absolute pressure sensors type 4045A2, 4075A50, and 4075A100. Experiments were conducted with equivalence ratios ranging between 0.7 and 1.3.

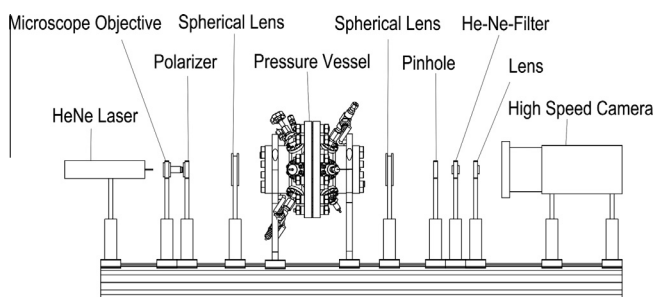


Fig. 1. Schematic of the experimental setup

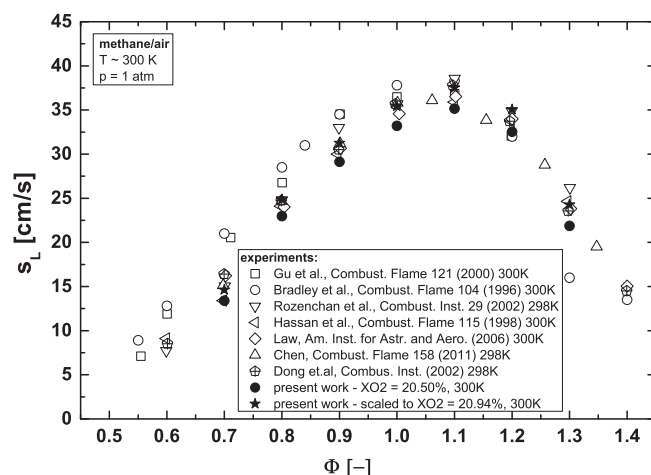


Fig. 2. Laminar burning velocities for methane/air flames as function of equivalence ratio; $p_i = 1 \text{ atm}$ and $T_i \sim 300 \text{ K}$; $X_{\text{O}_2} = [\text{O}_2]/([\text{O}_2] + [\text{N}_2])$.

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