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# Laminar flame speeds and ignition delay times of methane–air mixtures at elevated temperatures and pressures

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## HIGHLIGHTS

- The measured laminar flame speeds from the constant volume bomb and the counter flow flame are compared.
- GRI Mech 3.0, USC Mech II, and Aramco Mech 1.3 mechanisms are validated at elevated pressures.
- Overall reaction order are analyzed at initial pressures up to 6.0 MPa.
- Correlations for laminar burning velocities and ignition delay time of methane–air mixtures are provided.

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## ABSTRACT

Measurements on laminar flame speeds and ignition delay times of methane/air mixtures at elevated pressures and temperatures were carried out in a constant volume bomb and shock tube. The performances of GRI Mech 3.0, USC Mech II, and Aramco Mech 1.3 mechanisms were also evaluated from the data obtained. Results showed that the measured laminar flame speeds from the constant volume bomb by the linear method are slightly higher than those from the counter flow flame at rich mixtures and lower at lean mixtures. At rich mixtures, the laminar flame speeds with linear method are higher than that with non-linear method. The available mechanisms give slight overprediction to the constant volume bomb data at lean mixtures, and large underprediction at rich mixtures at elevated temperatures and pressures. Overall reaction order decreases and then increases with the rising of pressure from 0.1 to 10.0 MPa because of the chain reaction mechanism. For the ignition delay times, the three mechanisms are in good accordance with the experimental data of lean and stoichiometric mixtures at atmospheric pressure, while the discrepancy between calculation and measurement is increased at elevated pressures. These mechanisms seem to lack good sensitivity to the rich mixtures, especially at elevated pressures. Correlations for laminar burning velocities and ignition delay time of methane–air mixtures are provided.


## 1. Introduction

Since fossil fuel is depleting and automotive emission regulation is strengthening, researches are paying increasing attention on the study of alternative and clean fuels. One of the prospective alternative fuels is natural gas. Methane is the major constituent of natural gas as well as the smallest hydrocarbon fuel. Therefore, methane is a key fuel candidate of research. Methane has been in use on specific combustion devices like internal combustion engines and industrial gas turbines operated at high pressure and temperature.

Laminar flame speeds is a fundamental property of fuels, resulting from the combined influences of diffusivity, exothermicity, and reactivity. Besides, it is a key parameter in descripting complex combustion phenomena such as flame stabilization, extinction, turbulent flame structure and velocity [1–3]. Previously, extensive experiments have been conducted to measure the laminar burning velocities covering a wide range of conditions. The correlation of laminar flame speed as a function of temperature and pressure is necessary for the CFD simulation [4]. A plenty of research focused on the high dependence of methane–air mixture laminar flame speed on pressures and temperatures. Recently, Goswami [5] and Ranzi [6] summarized the experimental laminar burning velocities of methane–air mixtures at different pressures and temperatures for the latest sixty years, finding that large uncertainty still exists in the data. Furthermore, prediction of laminar flame speed with

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the GRI Mech 3.0 mechanism shows poor performance at high pressures and temperatures [7].

The methodologies for fundamental flame speed determination involve flames that are either stationary, or propagating with respect to a quiescent unburned mixture. The former includes conical, flat, and counter-flow flames and the latter refers to the spherically expanding flames. Recently, spherically expanding flame has been widely applied to measure the laminar burning velocities for a number of fuels. The stretch rate of spherically expanding flame is well defined and this method is the best choice for the measurement at higher pressures and temperatures. In this work, the spherical expanding flame was used to measure the laminar flame speeds at elevated pressures and temperatures.

Ignition delay time is also a key parameter of fuel chemistry, which can serve as the validation parameter in the development of chemical kinetics [8]. Shock tube is a standard facility to measure the ignition delay time at high temperatures. It is zero-dimensional and homogeneous inside, so the ignition of fuel oxidizer mixtures is controlled by chemical kinetic. Additionally, shock tube can measure the ignition delay time at the specified pressure and temperature. Although much research has been reported on the ignition delay time of methane/oxygen, most of the experiments were conducted under high argon dilution conditions [9–14], and only a few work reports the auto-ignition of methane–air mixtures [15,16]. Thus, measurement on ignition delay times of methane–air mixtures is still rare and worthwhile.

The objectives of this study are to measure the laminar burning velocities at various initial pressures and high temperatures (up to 443 K), to measure the ignition delay times of methane–air mixtures under various conditions, to evaluate the kinetic models on the basis of the measured data, to discuss the pressure effect on the chain reaction mechanism, and to provide the correlations for the laminar burning velocity and the ignition delay time of the methane–air mixtures.

## 2. Experimental and numerical methods

Recently, Egolfopoulos [17] reviewed different experimental approaches on determination of the laminar flame speed, and recommended the spherically expanding flame method when the pressure is greater than 0.5 MPa. Details of the experimentations in this study can be found in previous literatures [18,19], and here only a brief introduction is given. The spherically propagating flames are generated in a cylindrical combustion chamber (5.5 L) bearing pressure up to 10 MPa and initial temperature of 500 K by central ignition. The propagation of flame is then imaged with Schlieren photography and recorded by a high-speed digital camera (Phantom V611) operating at 10,000 frames per second, at 720 × 720 pixels, and magnification ratio of 0.11 mm/pixel.

Mixture preparation and the resulted uncertainty in equivalence ratio are described in the following. In the spherical vessel, all pipes, valves and vessel parts sensitive to fuel condensation are heated. Partial pressure is used to accurately measure and control the filling process. Partial pressure is measured with a high-accuracy pressure transmitter (Rosemount 3051). The absolute uncertainty in equivalence ratio is less than 0.0093. In addition, mixture preparation and the resulted uncertainty in the equivalence ratio of the experiments by other research groups [7] are provided in Table 1. It can be seen that the uncertainties in mixture preparation in this study is equivalent to those of others in the laminar flame speed measurement.

**Table 1**
Uncertainties in mixture preparation of other groups.

| Setup | Research group | Equivalence ratio uncertainty |
|---|---|---|
| Counter flow | USC | Less than 0.5% |
| Heat flux | TUE | Max ± 0.02 (absolute) |
| Spherical vessel | CORIA | Less than 0.01 (absolute) |
| | ICARE | Less than 0.0004 (absolute) |
| | PRISME | Max ± 0.76% |
| | RWTH | Less than 0.8% |

Post processing of the spherical flame data is obtained from the information of the expanding flame radius over time. When the flame maintains its stability, the flame propagation speed ($S_n$) can be extracted according to $S_n = dR/dt$, where $R$ is the recorded flame radius history. The existence of stretch in the front of the flame is due to its spherical shape. The flame stretch rate can be obtained from the equation $\alpha = 2S_n/R$. In this study, the linear and non-linear extrapolation methods are used to determinate the laminar flame speed. The linear method is that the flame propagation speed is linear to the stretch rate within a certain range in which the ignition effect and pressure rise are negligible; that is, $S_l - S_n = L_b \cdot \alpha$, where $S_l$ is the unstretched propagation speed and $L_b$ is the burned gas Markstein length. From mass conservation across the flame front, the unstretched laminar burning velocity ($S_u$) can be calculated by the formula $S_u = \rho_b/\rho_u \cdot S_l$, where $\rho_u$ and $\rho_b$ are respectively the unburned and burned gas densities. The non-linear extrapolation method is proposed by Kelly and Law [20]. By taking into account of the effect of ignition energy and pressure rise in the combustion chamber, flame photos in the range of 5 mm–25 mm are used in the analysis. In addition, we know that the flame front presented cellular instabilities at elevated pressures, so the data range were also restricted by the occurrence of cellular structure. The flame surface is smooth, free from any flame front diffusional-thermal and hydrodynamic instabilities.

A shock tube with an inner equivalent diameter of 11.5 cm was used to measure ignition delays. The detailed experimental setup has been presented and its validation has been conducted in reference [21]. The shock tube consists of a driver section and a driven section divided by double diaphragms, which are 4.0 m long and 4.8 m long respectively. Fuel mixtures entered into the driver section after the 12 hours' mixing in a cylindrical tank to ensure full homogeneity. Purities of methane, oxygen and argon are 99.9%, 99.999% and 99.999%, respectively. The measured ignition delay time ($\tau$) in this study is defined as the time interval between the time when an incident shock wave arrived at the endwall of the shock tube and the intercept of the maximum slope of $CH^*$ emission profile with the baseline.

Laminar flame speed was simulated with Premix code [22], which applies a hybrid time-integrating/Newton iteration method to solve the steady-state mass, species and energy conservation equations and can simulate the propagating burning process. TWOPNT, a boundary value problem solver in the Chemkin package [23], are used to solve equations. A transport property processor and a gas-phase interpreter which carry the species transport properties and process the chemical reaction mechanism are also built in the Chemkin package. Mixture-averaged transport properties are employed in this calculation. Multi-component transport option is preferred by certain modeling workgroups, such as Resources Research Institute and University of Leeds; while we choose the mixture-averaged transport properties like Lawrence Livermore National Laboratory and other groups. Ten continuation options are used in all calculations and the values of adaptive grid parameters (GRAD and CURV) varies from 0.99 to 0.01 for each case in order to obtain the grid-independent solutions. The final solutions (GRAD = 0.01, CURV = 0.01) were usually obtained with about 1300 mesh points. Convergence level for most cases is typically 2 cm $s^{-1}$ of final grid for calculating laminar flame speeds, which is accurate enough for our results.