



Full Length Article

Experimental and modeling study on ignition delay times of dimethoxy methane/*n*-heptane blends



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HIGHLIGHTS

- Ignition delay times of DMM/*n*-heptane blends were measured.
- A DMM/*n*-heptane blend model was developed.
- The effects of DMM addition on ignition delay times of DMM/*n*-heptane blends were analyzed.

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ABSTRACT

Ignition delay times of dimethoxy methane (DMM), *n*-heptane and DMM/*n*-heptane mixtures were measured behind reflected shock waves. The experiments were performed at the temperature ranges of 1100–1600 K, at pressures of 2 and 10 atm and equivalence ratios of 0.5, 1.0 and 2.0. A chemical kinetic model for DMM/*n*-heptane blends was built and validated with the experimental data. The calculations well agreed with the measurements for pure fuel and binary fuels. Based on the experimental and calculated results, effects of pressure, equivalence ratio and DMM blending ratio on ignition delay times were analyzed. Through sensitivity analysis, the most important reactions were found and then the effect of equivalence ratio on ignition delay times was interpreted. The ignition delay times of DMM/*n*-heptane blends decreased with the addition of DMM. Kinetic analysis revealed that the concentration of radical pool increased with the increase of DMM addition, resulting in an enhancement effect on the chemical reactivity.

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1. Introduction

As the growing air pollution caused by the worldwide use of fossil fuels for ground transportation, the oxygenated fuels, such as ethers, acetates, alcohols, furans, esters, carbonates, have been proposed as possible additives to conventional fossil fuels to reduce exhaust emissions. It has been proved that the addition of oxygenated fuels could significantly reduce the soot emissions in diesel engines [1–5].

Among these oxygenates, dimethoxy methane (DMM), also called methylal, has attracted the increasing attention as a pure fuel or blending fuels with diesel. DMM has the high oxygen content and no C–C bond in its molecular structure, which ensures soot-free combustion. Extensive engine tests have demonstrated the good emission-reducing characteristics of DMM [6–9]. Regarding fundamental combustion research, Daly et al. [10] investigated the oxidation of DMM in a jet-stirred reactor at 5.07 bar, 800–

1200 K, and equivalence ratios from 0.44 to 1.78. A detailed chemical kinetic model was developed, which agrees well with the experimental results. Renard et al. [11] studied the effect of DMM addition on the flame structure of ethylene flame, and the results showed a decrease of maximum mole fraction of soot precursors. The intermediate species in DMM diffusion flames were measured by Sinha and Thomson [12]. It was found that the absence of C–C bonds in DMM could effectively decrease the formation of the soot precursors. Curran et al. [13] studied the influence of the addition of oxygenated hydrocarbons to diesel fuels using a detailed chemical kinetic model which includes the sub-models of methanol ethanol, dimethyl ether, dimethoxymethane (DMM) and methyl butanoate. Dias et al. [14] extended an ethylene mechanism by building a sub-mechanism of DMM, and excellent agreements between experimental results and calculated mole fraction profiles for main species were observed. Chen et al. [15] studied the influences of different oxygenates (methanol, DMM, and DMC) addition to premixed *n*-heptane flames. The results showed that the concentrations of most C1–C5 hydrocarbon intermediates and benzene were reduced as oxygenated fuels were

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added. Zhang et al. [16] measured the ignition delay times of DMM in a shock tube at different initial conditions, and validate the chemical kinetic models in literatures. It is noted that the research objectives of the work of Zhang et al. [16] and this work are different. The work of Zhang et al. [16] aims at the mechanism validation and fundamental research of neat DMM ignition, while this work mainly focus on the development of a DMM/*n*-heptane blend model and the effect of DMM addition on the auto-ignition characteristics of *n*-heptane. The ignition delay time is crucial for HCCI engine control and chemical kinetic development. DMM addition to *n*-heptane can control the combustion phase of *n*-heptane HCCI engine and diesel engine. To our knowledge there is no study on the ignition of DMM/*n*-heptane blends in literature until now.

In this study, ignition delay times of DMM/*n*-heptane mixtures under various conditions were measured behind reflected shock waves. A DMM/*n*-heptane blending model was developed based on the experimental data. The sensitive analysis and radical analysis were conducted to interpret the ignition characteristics of DMM/*n*-heptane blends.

2. Experimental approach

All experimental research of this study was conducted in a shock tube, which has been described in details in previous publications [17,18]. A brief introduction about this apparatus was given as follows. The shock tube with a diameter of 11.5 cm is composed of three sections: the 2.0 m long driver section, the 5.3 m long driven section and the 0.6 m long flange section. The flange section is located between the driver and driven sections and separated by two polyethylene terephthalate diaphragms. Diaphragms of different thickness were selected according to the magnitude of the reflected pressure. The shock wave is generated by bursting the diaphragms. Mixtures of high-purity helium and nitrogen were used as the driver gas. The driven section needs to be evacuated to the pressure below 10^{-6} bar by a Nanguang vacuum system (ZJP-150 Roots vacuum pump and 2ZX-30D rotary vane vacuum pump) before each experiment, and then it was charged with the test mixtures which were prepared in a 128 L stainless steel tank. DMM and *n*-heptane were injected to the evacuated tank separately to their respective partial pressures which were kept below 50% of their saturated vapor pressures. Both oxygen and argon with purities over 99.999% are then charged to meet the desired equivalence ratio.

Four pressure transducer (PCB 113B26) were located along the driven section at fixed intervals (30 cm). The time intervals between the instants of shock wave arrival at each pressure transducer location were measured by three time counters (Fluke PM 6690) and then the shock velocity is calculated respectively. Based on these collected data, the speed of the endwall incident shock wave was determined by linear extrapolation of the axial velocity to the endwall. Another fast-response pressure transducer (PCB 113B03) was mounted at the endwall of the driven section to capture the endwall pressure signal which was regarded as the start point of the ignition delay time. The OH* emission signal was captured by a photomultiplier (Hamamatsu CR 131) together with a 307 nm narrow band pass filter. A digital recorder (Yokogawa, DL750) was used to record all the experimental data. Conditions behind the reflected shock was calculated using the software Gaseq [19]. According to the work of Petersen et al. [20], the uncertainty of the reflected temperature is primarily sourced from the uncertainty of the incident shock velocity, which is determined by the uncertainties in the distances between any two pressure transducers and the shock pass time recorded by the time-counters. A standard root-sum-squares (RSS) method used by

Petersen et al. [20] was adopted to determine the uncertainties of the reflected temperatures,

$$T_5 = \frac{T_1[2(\gamma - 1)M^2 + (3 - \gamma)][(3\gamma - 1)M^2 - 2(\gamma - 1)]}{(\gamma + 1)^2 M^2} = AM^2 + B + CM^{-2} \quad (1)$$

$$M = \frac{V_s}{\sqrt{\gamma RT_1}} \quad (2)$$

$$\delta V_s = \sqrt{\left(\frac{1}{\Delta t} \delta \Delta z\right)^2 + \left(\frac{-\Delta z}{\Delta t^2} \delta \Delta t\right)^2} \quad (3)$$

$$\delta T_5 = \frac{\partial T_5}{\partial M} \delta M = (2AM - 2CM^{-3}) \frac{\delta V_s}{\sqrt{\gamma_1 RT_1}} \quad (4)$$

where the T_5 , T_1 , γ , V_s , R denote the reflected temperature (K), initial temperature (K), adiabatic exponent, velocity of the incident shock wave (m/s), the universal gas constant, respectively.

The uncertainty of the time interval detected by the FLUKE PM6690 time-counter was estimated to be 1000 ns mainly due to the resolution time of the pressure transducer. The revolution of the shock pressure transducer, the sensing area of the transducer and the shock front thickness are primarily attributable to the uncertainty of the distance. In this study, the sum total of distance uncertainties induced by the above-mentioned three factors was estimated to be $2 \times 1000 \text{ ns} \times V_s$. Adopting the RRS method, the largest uncertainties of the reflected temperature was estimated to be 20.5 K. In this study, the ignition delay time error comes mainly from the uncertainty of the temperature and the pressure. Ultimately, these two factors lead to an uncertainty of within 19% in all measured ignition data, τ_{ign} , based on the Arrhenius type correlation of τ_{ign} and using the RSS method. A detailed description can be found in the work of Zhang et al. [21].

The definition of the ignition delay is the time interval between the arrival of the shock wave at the endwall and the extrapolation of the steepest rise in the captured OH* concentration profile to the baseline, as shown in Fig. 1. It is noted that a certain pressure rise ($dp/dt = 4\%/ms$) can be observed before ignition due to the boundary effect, and this has been taken into account in the numerical simulations. Fig. 2 depicts the comparison of the ignition delay times measured in this study with those investigated by Horning et al. [22], as well as the simulated results using the LLNL *n*-heptane model [23,24]. The good agreement between the

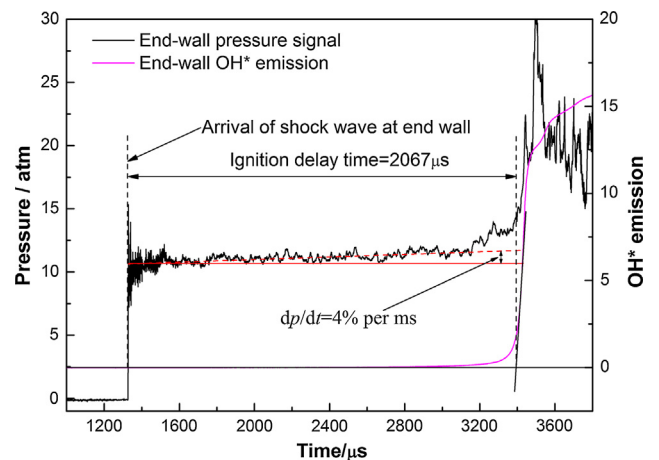


Fig. 1. Typical endwall pressure and OH* chemiluminescence signal of DMM/O₂/Ar mixture, at $p = 10.0$ atm, $T = 1113$ K, and $\phi = 1.0$.

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