



Ignition delay times of low-vapor-pressure fuels measured using an aerosol shock tube

Daniel R. Haylett^{a,*}, David F. Davidson^b, Ronald K. Hanson^b

^aLawrence Livermore National Laboratory, Livermore, CA 94550, USA

^bHigh Temperature Gasdynamics Laboratory, Department of Mechanical Engineering, Stanford University, Stanford, CA 94305, USA

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ABSTRACT

Gas-phase ignition delay times were measured behind reflected shock waves for a wide variety of low-vapor-pressure fuels. These gas-phase measurements, without the added convolution with evaporation times, were made possible by using an aerosol shock tube. The fuels studied include three large normal alkanes, *n*-decane, *n*-dodecane and *n*-hexadecane; one large methyl ester, methyl decanoate; and several diesel fuels, DF-2, with a range of cetane indices from 42 to 55. The reflected shock conditions of the experiments covered temperatures from 838 to 1381 K, pressures from 1.71 to 8.63 atm, oxygen concentrations from 1 to 21%, and equivalence ratios from 0.1 to 2. Ignition delay times were measured using sidewall pressure, IR laser absorption by fuel at 3.39 μm , and CH^* and OH^* emission. Measurements are compared to previous studies using heated shock tubes and current models. Model simulations show similar trends to the current measurement except in the case of *n*-dodecane/21% O_2 /argon experiments. At higher temperatures, e.g. 1250 K, the measured ignition delay times for these mixtures are significantly longer in lean mixtures than in rich mixtures; current models predict the opposite trend. As well, the current measurements show significantly shorter ignition delay times for rich mixtures than the model predictions.

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1. Introduction

There is a need to extend the study of purely gas-phase combustion chemistry to include low-vapor-pressure fuels. Conventional shock tubes use gas-phase mixtures and can provide, behind the reflected shock wave, ideal, nearly zero-dimensional, nearly constant-volume reactor conditions up to the time of ignition, or longer in highly dilute mixtures [1]. However, this conventional operation limits the fuels that can be easily loaded into the shock tube to gases and liquids with a sufficiently high vapor pressure. Heating the shock tube and mixing manifold can raise the vapor pressure to allow the study of heavier fuels [2], but this process can be limited by pre-test fuel decomposition or oxidation. To avoid these issues we have developed a method of injecting aerosols into the shock tube and evaporating them behind the incident shock wave, thus producing a purely gas-phase test mixture for study behind reflected shock waves [3,4]. Using this method, the range of fuels that can be studied is greatly extended, and it is now possible to investigate the ignition kinetics of practical heavy fuels that are used industrially, as well as their kinetically-simpler surrogates.

Ignition delay times are good indicators of the overall behavior of combustion reactions and are regularly used as performance benchmarks for detailed chemical mechanisms. Validation of the detailed chemical mechanisms is very important, because these mechanisms are obliged to use many reactions and rate constants that are only estimated or theoretically predicted and not derived directly from experiment. For large fuel molecules (which typically have low vapor pressures), the number of species that are formed during decomposition and oxidation can be very large. For example, a Lawrence Livermore National Labs (LLNL) chemical mechanism for alkanes up to *n*-hexadecane ($\text{C}_{16}\text{H}_{34}$) contains over 2000 species and over 8000 reactions [5]. Ignition delay times provide useful targets and some constraint on the modeling of these complex systems.

Much work is still needed to fully understand the combustion of large fuel molecules, because of the increased complexity of their detailed mechanisms and the added difficulty in performing experiments with these fuels [18]. This is unfortunate, because most of the devices that utilize combustion are powered by low-vapor-pressure fuels including diesel and bio-diesel fuels, and rocket and jet fuels. Many new mechanisms have been developed recently [5–9,25,28], but more experimental validation targets are required for these fuels and their surrogates [27]. To help address this need, we have initiated studies of a variety of low-vapor-pressure fuels

* Corresponding author.

E-mail address: danhaylett@gmail.com (D.R. Haylett).

using an aerosol shock tube methodology, providing in many cases, the first purely gas-phase shock tube ignition delay time measurements for these fuels.

2. Low-vapor-pressure fuels

Fuels used in combustion devices are rarely composed of a pure single component, and in order to model a real, multi-component fuel, single- or multi-component surrogate mixtures are often employed [10,27]. This motivates the study of various types of pure fuels that display characteristics similar to real fuels, in addition to the study of the real fuels themselves. Because fuels such as jet fuel, diesel, and bio-diesel are non-volatile, the surrogate components that best represent these fuels also have low-vapor-pressures, and the aerosol shock tube is well suited for the study of these fuels. Here we focus on *n*-alkanes, which can make up a significant fraction of distillate diesel fuels themselves, and methyl decanoate as a large methyl ester that is structurally related to bio-diesel.

2.1. *n*-Decane

There have been many ignition delay time studies of *n*-decane [11–14,24,26]. This is in part because its vapor pressure at room temperature is near 1.4 torr, which generally is sufficient to make a purely gas-phase mixture at relevant mixture fractions, with the exception of experiments at high pressures. *n*-Decane ($C_{10}H_{22}$) has been commonly used as the alkane representation for jet fuel-relevant surrogates, but has also been used for diesel [13] and bio-diesel [8] comparisons.

2.2. *n*-Dodecane

Less abundant are ignition delay time studies focused on *n*-dodecane [11,15]. The vapor-pressure at room temperature of *n*-dodecane is only 0.13 torr. This limits the fuel loadings that can be achieved, and hence heating is usually necessary to perform shock tube experiments. *n*-Dodecane ($C_{12}H_{26}$) has also been used as the alkane component in jet fuel surrogates.

2.3. *n*-Hexadecane

There are very few studies on the purely gas-phase oxidation of *n*-hexadecane [6,16,17] because its vapor pressure at room tem-

perature is only 1.4 mtorr. *n*-Hexadecane (cetane) is a major alkane constituent of diesel fuel, and appears to be very important in influencing its behavior upon oxidation. Also, *n*-hexadecane ($C_{16}H_{34}$) is one of the species that is used in the definition of the cetane number scale to characterize the combustion characteristics of diesel fuel. By definition *n*-hexadecane has a cetane number of 100.

2.4. Methyl decanoate

The authors are unaware of any purely gas-phase shock tube studies of methyl decanoate, despite the fact that it is a important surrogate fuel for the study of methyl ester based bio-diesels. A detailed mechanism has been developed at LLNL that predicts the behavior of methyl decanoate oxidation [8].

2.5. Diesel (DF-2)

Diesel fuel is a low-volatility distillate fuel widely used in combustion devices. Heated shock tubes have been used recently in the past to study diesel ignition [19,20], but with a distillation curve that extends up to 350 °C (623 K), it can be a very difficult task to get all of the heavier components of the fuel mixture into the shock tube. Here we examine several different diesel fuel samples to show how variability in composition and cetane index affect ignition delay times.

3. Experimental setup

Our current aerosol shock tube methodology is described in Haylett et al. [3,4]. The aerosol shock tube and supporting diagnostics are shown in Fig. 1. A series of fast-response pressure sensors are used for measuring the incident shock speed. A mid-IR (3.39 μ m) laser and a visible (660–670 nm) laser are used for measuring fuel loading. A Kistler 601B piezo-electric transducer located near the endwall (3 cm) is used as the primary measure of ignition delay time. CH^* (at 431 nm) and OH^* (at 306 nm) are both detected through sidewall windows located near the endwall, also to provide ignition delay data.

The aerosol is first created in a mixing tank connected to the endwall of the shock tube, using a bank of ultrasonic nebulizers, and introduced through an endwall gate valve. The aerosol is slowly pulled into the test section (by a vacuum line located near the upstream gate valve) to ensure uniformity of the test mixture.

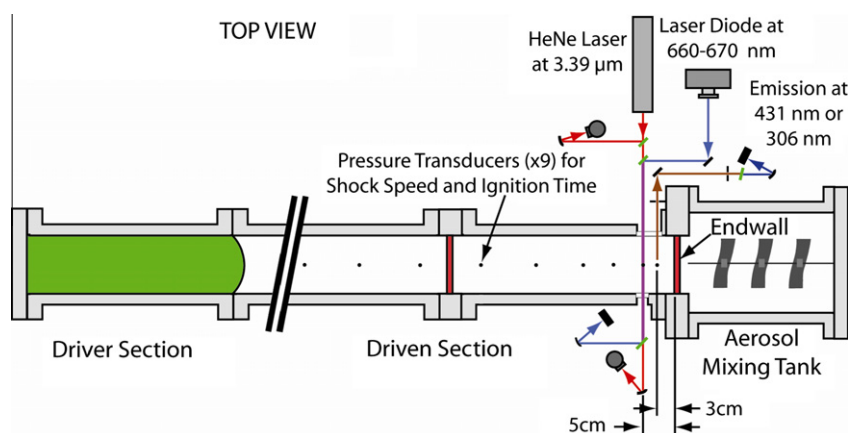


Fig. 1. Schematic of aerosol shock tube with pressure, emission, and laser diagnostics. The pressure sensors are used for shock speed measurement and ignition delay time determination, the mid-IR HeNe laser is used for absorption-based fuel measurements, the visible laser diode is used for droplet scattering measurements, and the emission measurement is used to measure ignition delay time. Sliding gate valves are shown in red as the shock tube end wall and 1 m from the end of the end wall. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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