



A shock tube ignition delay study of conventional diesel fuel and hydroprocessed renewable diesel fuel from algal oil



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HIGHLIGHTS

- Ignition delay measurements reported for conventional and alternative diesels.
- Ignition delay indistinguishable for $T > 1000$ K, varies substantially for $T < 1000$ K.
- Empirical model is presented for ignition variability.

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ABSTRACT

Ignition delay time measurements were made using the shock tube method for a conventional petroleum-derived military diesel fuel, F-76, and an alternative hydroprocessed renewable diesel fuel derived from hydroprocessing algal oils, HRD-76. Measurements were made in the reflected shock region using pressure and chemiluminescence and carried out for temperatures ranging from 671 to 1266 K, at nominal pressures of 10 and 20 atm, and for fuel–air equivalence ratios of 0.5 and 1.0. The measurements show that the high-temperature (>1000 K) reactivity of the two fuels is indistinguishable and very similar to that for petroleum-derived and alternative jet fuels and biodiesels. While at low temperatures (<1000 K) the ignition delay time for HRD-76 is up to a factor of two shorter than F-76 due to the larger fraction of n-paraffins and lack of aromatics in HRD-76. The combination of the present diesel ignition measurements with previous studies for jet fuel ignition illustrate common characteristics for the gas-phase ignition of these high molecular weight mostly-aliphatic fuels, notably common high-temperature ignition characteristics and low-temperature reactivity that correlates with derived cetane number (DCN). These characteristics allow for formulation of a simple three-Arrhenius model for ignition delay time that incorporates DCN functionality to describe variability in fuel reactivity.

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1. Introduction

Worldwide consumption of petroleum distillate fuel oil, which includes diesel fuels and fuel oils, has been around 25 million barrels per day since 2007–2008 [1] where it accounts for about 10% of worldwide energy consumption. Diesel fuels are primarily used in transportation and power-generation where they are most often burned in diesel engines. The prediction of autoignition, which initiates combustion in diesel and other compression-ignition engines, is of critical importance to the computational design and optimization of diesel engines, today largely based on computational fluid dynamics calculations that include fuel chemistry. Additionally, many alternative diesel fuels have been developed

in recent years, including biodiesels from plant oils, animal fats, and waste oils; synthetic fuels from gas-to-liquid and coal-to-liquid processes; synthetic fuels from hydroprocessing bio-oils; and fuels from genetically engineered microbes. These fuels can have different physical and chemical kinetic properties, in some cases unknown, which can influence their use in engines.

Most experimental work on the autoignition of diesel fuels found in the literature is based on the autoignition of fuel sprays in constant volume chambers [2], engines [3], or rapid compression machines [4]. While there have been many studies of the gas-phase autoignition of diesel fuel surrogate compounds [5–8], typically smaller compounds (e.g., n-heptane), very few experiments have been performed where the gas-phase autoignition of multi-component diesel fuels has been isolated from evaporation and mixing phenomena, due to the low volatility of diesel fuels. This void in the literature is problematic due to the importance of low-volatility fuels in the transportation and energy sectors and

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the growing number of kinetic models (see reviews [9,10]) that lack sufficient targets for validation.

Here we present measurements of ignition delay times for conventional and alternative diesel fuels carried out using the reflected shock technique, which provides well defined reaction conditions of relevance to combustion engine applications. Prior shock tube studies of gas-phase multi-component petroleum diesel fuel autoignition are limited, to our knowledge they only include the work Haylett et al. [11] and Penyazkov et al. [12]. Haylett et al. [11] measured ignition delay times for four diesel fuels at 3–7 atm, temperatures from 950 to 1400 K, and for mixtures containing 4% and 21% O₂ in argon with highly variable fuel loading. Haylett et al. used an aerosol shock tube technique where the fuel was introduced into the shock tube as an aerosol, the incident shock was used to vaporize the fuel, and measurements were made behind the reflected shock. Penyazkov et al. [12] used a heated shock tube method, similar to that utilized here, to measure ignition delay times for a Diesel fuel No. 2 (DF-2) in air at 5–10 atm, 1065–1838 K, and for equivalence ratios of 0.5, 1, and 2.

There has been important recent progress towards developing synthetic non-oxygenated alternative diesel and jet fuels from renewable biomass sources using hydroprocessing to convert bio-oils to paraffinic hydrocarbons. These fuels have been labeled as hydroprocessed renewable diesel or jet (HRD or HRJ) fuels and have been demonstrated in application (e.g., the US Navy 2012 Rim of the Pacific demonstration exercise in anticipation of the “Great Green Fleet” [13]) and in some cases certified [14] for use in diesel and gas turbine engines. Due to their largely straight chain and branched paraffinic composition, HRD and HRJ fuels can have different combustion properties than petroleum fuels containing appreciable quantities of aromatics and cyclo-alkanes with lower reactivity, higher density, and other differing properties. One important observation has been the higher reactivity of HRJ fuels relative to petroleum jet fuels [15]. Here we present a comparison of the ignition delay time, a measure of global reactivity, for a conventional military diesel fuel, F-76, and a HRD fuel synthesized from algal oil, HRD-76.

2. Experimental method

Ignition delay time measurements were carried out in the Rensselaer heated shock tube facility (5.7 cm inner diameter, 2.59 m long driver, 4.11 m long driven) using the reflected shock technique. The facility and experimental methods for ignition delay time measurements have been previously described [16,17]. Here only details pertinent to the present measurements are given.

Due to the high molecular weight of the diesel fuels studied, the shock tube and mixing vessel were heated to 200 °C to provide sufficient fuel volatility for gas-phase fuel/air mixture preparation (air is 99.995% pure O₂ and N₂ at a ratio of 1–3.76) and avoid fuel condensation during shock tube experiments. The heated shock tube exhibited axial temperature uniformity of ±4 K during the experiments. Gas chromatography/mass spectrometry measurements (Shimadzu QP5050) were made to ensure that the diesel fuels were not partially decomposed at the 200 °C mixture preparation condition.

Experiments were carried out for two diesel fuels: F-76, a military grade petroleum-derived diesel fuel, and HRD-76, an alternative diesel fuel from hydroprocessed algal oil. Chromatograms and the hydrocarbon class distribution for the two fuels are given in Figs. 1 and 2, respectively. Some fuel properties are given in Table 1, while additional detailed fuel data sheets are appended as supplementary material. The experimental conditions for which ignition delay time measurements were carried out are given in Table 2. Measurements at the lowest temperatures were limited to lean

conditions ($\phi = 0.5$), requiring less fuel loading (lower temperature shock tube experiments require higher initial total pressure), due to the relatively low volatility of the fuels studied. It can be seen that F-76 contains cyclo-alkanes, aromatics, and other compounds not found in HRD-76, which is wholly comprised of normal and lightly-branched alkanes. These compositional differences result in a substantially higher derived cetane number (DCN measurements found in Table 1, carried out by Advanced Engine Technology Ltd. using ASTM D6890) for HRD-76 relative to F-76. F-76 also contains a broader range of molecular weights, as shown by the chromatograms. For the purposes of defining fuel/air mixture stoichiometry, the average molecular formula (C_mH_y in Table 1) was defined using a correlation between distillation curve and molecular weight from Maxwell and Bonnell [18] and the hydrogen-to-carbon ratio (H/C Table 1, hydrogen content determined using ASTM D7171).

For the determination of ignition delay times behind reflected shock waves, pressure and electronically-excited OH chemiluminescence were measured. See Fig. 3 for an example ignition delay time measurement. Ignition delay time was defined as the time interval between shock reflection at the driven section end wall, defined using the measured pressure profile at a side wall location (2 cm from end wall) and the measured incident shock velocity, and the onset of ignition at the end wall, defined by extrapolating the maximum slope in the OH signal to the baseline (acquired through the shock tube end wall). The pressure gradient behind the reflected shock wave, due to viscous gasdynamics, for these experiments was $(dp/dt)(1/P_0) = 2\text{--}3\% \text{ ms}^{-1}$. This pressure gradient should be considered in kinetic modeling of these experiments, particularly for ignition delay times longer than 2 ms which could be reduced by tens to near one hundred percent compared to ignition delay times predicted for constant volume conditions, depending on the conditions and model.

Reflected shock conditions were determined using the normal shock equations with the measured shock velocity and reactant mixture thermodynamic properties as inputs. Thermodynamic surrogates were used to represent F-76 and HRD-76 for the defining reactant thermodynamic properties and were formulated to emulate the fuel H/C, average molecular weight, and organic distribution given in Fig. 2, as in [16]. The thermodynamic surrogate for the fuel has minimal influence on the calculated reflected shock condition because the reactant thermodynamic properties are dominated by the ~99% air contained in the fuel/air mixtures. For example, the difference in reflected shock temperature for a calculation carried out using a thermodynamic surrogate, as described above, and a calculation carried out assuming a pure n-alkane fuel, with both calculations containing the same carbon content (that of the experiment), is approximately 1 K.

All measured ignition delay times are reported in the appended supplementary material. Ignition delay times have uncertainties estimated at ±25% based on uncertainties in determination of ignition delay time from measured signals and the uncertainties in reactant mixture composition and reflected shock conditions. The uncertainty in reflected shock temperature (±1–1.5%), based on uncertainty in incident shock velocity, has the largest contribution to ignition delay time uncertainty.

3. Results and discussion

Measurements of ignition delay times are shown in Fig. 4 for F-76 and HRD-76. In both cases the high-temperature results (>1000 K) exhibit an expected decrease in ignition delay time with increasing equivalence ratio and pressure. The $\phi = 0.5$ fuel/air results at 20 atm demonstrate the three regions of reactivity characteristic of aliphatic fuels: the high-temperature region, exhibiting

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