



Full Length Article

Ignition delay time correlations for distillate fuels



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ABSTRACT

Ignition delay times were measured behind reflected shock waves in a shock tube for a wide variety of distillate fuels over a range of temperatures, pressures and mixtures. The fuels studied include: jet fuels (JP-5, JP-8, and Jet A), rocket propellants (RP-2), diesel fuels (F-76 and DF-2) and gasoline. A simple correlation was found to describe the ignition delay times for all these fuel/air experiments for equivalence ratios near unity, temperatures from 1000 to 1400 K, and pressures from 6 to 60 atm. A simple correlation was also found for low-fuel-concentration experiments diluted in argon. Previously published ignition delay time data were found to be in good agreement with these correlations. Finally, for several fuels studied, systematic variations were seen in the activation energy for ignition delay time measurements with varying equivalence ratio and oxygen concentration.

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1. Introduction

The quality and composition of distillate fuels throughout the world varies significantly. With this variation comes a concern that engine performance will be critically affected by fuels with widely varying composition. For certain engines such as gas turbines, significant changes in ignition delay times, for example, can have a significant impact on engine operation. Similarly, high-performance spark ignition engines are sensitive to fuel composition and ignition delay time. For other engines, such as rockets, the assumption that “mixed is burnt” means that for the bulk combustion volume, chemical kinetics are not assumed to play a critical role. Chemical kinetics can however play an important role in predicting rocket engine oscillations and stability. In military engines, the “One Fuel” directive implemented by the U.S. Department of Defense, requires an understanding of the influence of using this one fuel, i.e. jet fuel, in engines designed for other fuels, such as diesel, further motivating the need to understand variations in ignition delay time across a range of distillate fuels.

Thus, there continues to be a wide research effort to develop accurate models for the combustion kinetics of jet fuels and other distillate fuels with particular interest on the role and importance of fuel composition. Critical to this development is a need for accurate kinetics target data to validate and refine these models. Common laboratory modeling targets include ignition delay time (IDT)

and flame speed, which place global constraints on model predictions, and species time-histories that place more specific constraints on the internal sub-mechanisms of these models. Although a wide literature on ignition delay time measurements in shock tubes exists, these data are primarily for neat hydrocarbons and to a much lesser extent for a range of practical distillate fuels. For a review of methods and data, for example, see Lifshitz [1] and Davidson and Hanson [2].

One concern of model developers is that because distillate fuels have variable compositions, kinetics models and fuel surrogates may have to be tuned to the specific composition of the individual fuel to accurately reproduce critical combustion parameter such as ignition delay times and flame speeds. Based on our current knowledge, for example of the large variation of ignition delay times for archetypal individual surrogate components, this concern is justified [3].

Distillate fuels, however, are not composed of a small and limited number of components as are surrogate fuels. Surrogate fuels are often designed to include a small number of selected individual archetypal components of *n*-alkane, iso-alkane, cyclo-alkane and aromatic species. GCxGC analyses of distillate fuels, on the other hand, demonstrate an immense array of different component species. While this large variability in composition might be viewed with concern, recent measurements of distillate fuel pyrolysis products have demonstrated that only a small number of common intermediate species form [4], providing the basis for a possible argument that some combustion characteristics of distillate fuels should actually be similar.

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Related to this, recent work on model development for practical distillate fuels (e.g. the hybrid model of Wang et al. [5]) assumes that ignition may be viewed at high temperatures, simply yet accurately, as initial pyrolysis (including abstraction reactions) followed by oxidation of stable intermediates. This assumption is completely consistent with shock tube measurements of the species time-history of fuel and intermediate fuel product species (e.g. the major product C_2H_4) during *n*-heptane, *n*-dodecane and *n*-hexadecane oxidation [6–8]. With this model, the observed commonality in decomposition products may work in the modeler's favor to simplify the task of generating a common mechanism for all jet fuels. In short, if most fuels generate approximately the same decomposition products, should not the ignition delay times be similar?

This question has been difficult to study because of the paucity of high-temperature IDT data for distillate fuels. In part, this is because of a lack of a single source for research fuel and the inherent variability of the generic fuel composition. Gauthier et al. measured ignition delay times in gasoline in 2004 [9]. Early jet fuel studies were performed by Dean et al. [10]. Vasu et al. in our laboratory then investigated IDT for a series of jet fuels [11]; this work was continued by Zhu et al. for F-76 [12], and Haylett et al. for DF-2 [13]. Wang and Oehlschlaeger [14] and Dooley et al. [15] provided high-pressure ignition delay time for a specific Jet A fuel (POSF4658). Liang et al. measured ignition delay times for China #3 aviation kerosene [16]; Steil et al. measured highly dilute kerosene in O_2 /argon [17]. Other workers have studied jet fuel ignition delay times, but without a major emphasis on gas-phase high temperature IDT (see Ref. [11]).

Recently, Gowdagiri et al. found in a study of F-76 and bio-fuels that high-molecular weight, mostly aliphatic fuels had very similar high-temperature ignition properties [18]. In their study, they found an IDT correlation for these fuels at temperatures above 1000 K of the form

$$\tau_{\text{ign}} = 1.7 \times 10^{-8} \phi^{-0.36} p^{-0.71} \exp(25.82/R[\text{kcal/mol K}]) \text{ [s]} \quad (1)$$

This expression captures the weak dependence on equivalence ratio (ϕ) and an approximate expected dependence on pressure p in atm and temperature T in K for these distillate fuels.

In this study, we further investigate the observation of Gowdagiri et al. about high-molecular-weight fuels and measure the ignition delay times for a wide variety of distillate fuels over a wide range of temperatures, pressures, and mixtures. We find that under certain constraints, these ignition delay times can all be well-correlated with simple relationships.

2. Experimental method

Ignition delay times were measured behind reflected shock waves in two shock tubes. High-pressure measurements (greater than 10 atm) were made in a heated 5.0 cm inner diameter shock tube using scribed aluminum diaphragms; low-pressure measurements (near 1.2 atm) were made in a heated 14.1 cm inner diameter shock tube using polycarbonate diaphragms. The experimental setup for the high-pressure shock tube is shown in Fig. 1. Both shock tubes are helium driven, turbo-molecular pumped, and heated to 90 °C; their accompanying gas mixing assemblies were also heated to 90 °C and turbo-molecular pumped. Full evaporation of the fuel components was verified visually by first evaporating small liquid samples of the fuel in an external glass volume at the same temperature and then expanding this gas into the mixing assembly at an even lower pressure. Separate tests were performed to ensure that the partial pressure in the mixing assembly scaled linearly with the liquid sample volume over the range of fuel sample volumes used for each experiment (typically 0.5–2 cc). Mano-

metric measurements (in the mixing assembly) and laser absorption measurements (in the shock tube) of the fuel loading were then compared to ensure that condensation losses in the transfer and shock tube filling processes were negligible. Incident shock speeds in both shock tubes were measured using arrival times from a series of 5 PCB™ pressure transducers approximately equally-spaced and located near the end wall. Reflected-shock temperatures and pressures based on the measured shock speed extrapolated to the end wall and the ideal frozen-chemistry shock relations, assuming vibrationally-equilibrated test gas in both the incident and reflected regions, were accurately determined ($2\sigma = 1\%$). The assumption that the reflected shock conditions are vibrationally equilibrated was tested and confirmed by Gauthier et al. in heptane/air mixtures [9]. Non-reactive pressure profiles in these experiments (using N_2 instead of O_2 in the mixture) were adjusted using driver inserts to limit pressure (and hence also temperature) variations to less than 1% over the required test times [19]. (A representative non-reactive pressure profile is shown in Fig. 2.)

Ignition delay times were measured using OH^* emission near 306 nm (Thorlabs™ model PDA36A photodiode, UG5 Schott™ glass filter, and simple collection optics) and side-wall pressure using a Kistler™ 603B PZT. Ignition delay times were defined as the time from the arrival of the reflected shock wave at the side-wall observation port (2 cm from the end wall for the low-pressure shock tube and 1 cm from the end wall for the high-pressure shock tube) to the time derived from back extrapolating the steepest rise in the OH^* or side-wall pressure signal to the local baseline. Ignition delay times using the two methods were normally in close agreement (within $\pm 3\%$) with each other.

The largest uncertainty in the IDT measurements can be associated with the influence of the reflected shock temperature on the ignition delay times. With activation energies for the majority of the experiments performed being near 40 kcal/mol, uncertainties of 1% in T_5 , the reflected shock temperature, translates into an absolute uncertainty in the ignition delay times of $\pm 15\%$. (Error bars are not shown on the figures as they are approximately the same dimensions as the symbols.) However, systematic differences in the data of 10% (e.g. because of pressure or fuel/oxygen loading) can readily be distinguished. Representative data for JP-8 and Jet A are shown in Fig. 2. In the JP-8 example, the ramp in 3.39 μm absorbance seen in the incident shock regime from -50 to $0 \mu\text{s}$ is a result of a small transverse misalignment of this beam across the shock tube. The temporally-broad pressure rise seen from -10 to $30 \mu\text{s}$ is a result of a bifurcation of the reflected shock wave traveling through the boundary layer that is seen with diatomic gases. The pressure rise seen near 200 ms before the ignition event at 210 ms is a result of weak non-local ignition increasing the local pressure. Individual experiments were examined for non-local ignition occurring away from the end wall and deflagration-to-detonation transitions (DDT) that can shorten measured IDT. Measurements where there is evidence of significant shortening of the measured IDT were not included in the dataset. IDT can also be estimated from rates of fuel consumption measured using IR laser absorption (see next paragraph). Measurements where the effects of non-local ignition or DDT start to become evident at the same time (within $\sim 5\%$) as the estimated IDT (from IR laser absorption) are deemed acceptable.

In order to minimize uncertainty in fuel concentration, this quantity was measured directly in the shock tube using IR laser absorption at 3.39 μm and Beer's law; absorption coefficients for each fuel that were measured in separate experiments in our laboratory using the method of Klingbeil et al. [20]. Typical uncertainties in the fuel loading based on these absorption coefficients have a Standard Error of $2\sigma = 2\text{--}5\%$. For all fuels tested, $C_N H_M$ values, needed for accurately determining gas-phase equivalence ratios

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