



Autoignition characteristics of oxygenated gasolines



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ABSTRACT

Gasoline anti-knock quality, defined by the research and motor octane numbers (RON and MON), is important for increasing spark ignition (SI) engine efficiency. Gasoline knock resistance can be increased using a number of blending components. For over two decades, ethanol has become a popular anti-knock blending agent with gasoline fuels due to its production from bio-derived resources. This work explores the oxidation behavior of two oxygenated certification gasoline fuels and the variation of fuel reactivity with molecular composition. Ignition delay times of Haltermann (RON = 91) and Coryton (RON = 97.5) gasolines have been measured in a high-pressure shock tube and in a rapid compression machine at three pressures of 10, 20 and 40 bar, at equivalence ratios of $\varphi = 0.45, 0.9$ and 1.8, and in the temperature range of 650–1250 K. The results indicate that the effects of fuel octane number and fuel composition on ignition characteristics are strongest in the intermediate temperature (negative temperature coefficient) region. To simulate the reactivity of these gasolines, three kinds of surrogates, consisting of three, four and eight components, are proposed and compared with the gasoline ignition delay times. It is shown that more complex surrogate mixtures are needed to emulate the reactivity of gasoline with higher octane sensitivity ($S = \text{RON} - \text{MON}$). Detailed kinetic analyses are performed to illustrate the dependence of gasoline ignition delay times on fuel composition and, in particular, on ethanol content.

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1. Introduction

Many advanced combustion engine technologies have been proposed in recent years to achieve higher efficiencies and lower emissions. Most of these technologies revolve around low-temperature combustion (LTC) concepts and include variants such as homogeneous charge compression ignition (HCCI), partially premixed combustion (PPC) and reactivity controlled compression ignition (RCCI) [1–3]. New combustion modes used for advanced combustion engines are mainly controlled by the autoignition of fuel/air mixture. Therefore, fundamental understanding of the chemical kinetics of fuel combustion is essential in the development and optimization of these new engine technologies [4].

Gasolines, one of the most widely used light duty automotive fuels, are complex mixtures containing hundreds of different chemical compounds such as alkanes, aromatics, naphthenes and

olefins. Thus, to develop a detailed reaction mechanism comprising all of the components present in gasoline in order to predict gasoline ignition behavior is intractable. Owing to this complexity, a popular approach to describe the combustion behavior of gasoline is to use a surrogate fuel composed of several reference components. Various gasoline surrogates, ranging from single to multi-components, have been proposed in the literature [5–8]. The primary reference fuels (PRFs), which are binary blends of *n*-heptane and *iso*-octane, are widely adopted, given their relatively well-validated chemistry. However, previous studies have shown that PRFs may not be suitable surrogates over wide range of engine operating conditions [8–11]. The physical and chemical properties of PRFs are different from those of real gasoline fuels because they not only contain alkanes and *iso*-alkanes but also cycloalkanes, aromatics, alkenes and oxygenates [9–13]. Several recent studies have instead proposed a ternary mixture of PRF/toluene (also known as toluene primary reference fuels, TPRFs) as gasoline surrogates [14–18]. By adding toluene to PRFs, TPRFs possess gasoline's inherent fuel sensitivity and have gone some way in assisting the development of simple gasoline surrogate models which capture important combustion metrics [16–18] such as ignition delay time and flame

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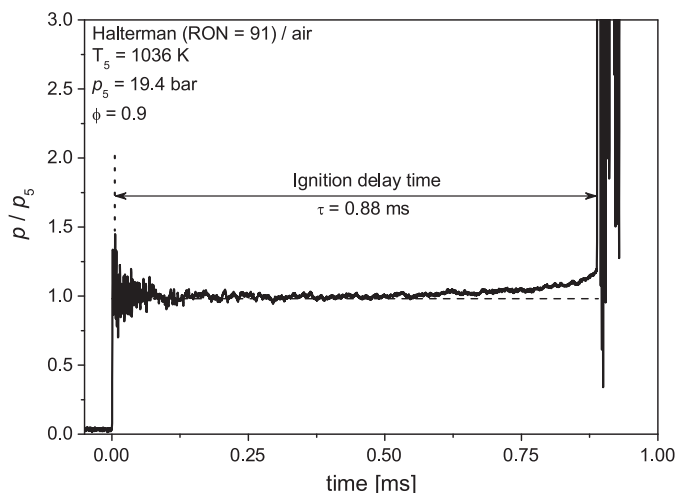


Fig. 1. Example gasoline/air pressure profile showing the definition of ignition delay time for shock tube experiments.

speed. With the availability of complex reaction mechanisms for many different alkenes [19] and cycloalkanes [20], four- and five-component surrogates containing 1-hexene and 1-pentene representing alkenes and methylcyclohexane representing cycloalkanes, respectively, have also been proposed [21–24]. Recently, Sarathy et al. [25] and Ahmed et al. [26] proposed multi-component surrogates to reproduce the low-temperature auto-ignition behavior of transportation gasoline fuels. In addition to various hydrocarbon classes present in gasoline fuels, the employment of oxygenated fuels such as ethanol in liquid transportation fuels has grown significantly in recent years. In various countries across the world such as Germany, France, Finland and in the U.S., ethanol is used as a blend of 10% by volume (E10) and the ethanol content in blends is anticipated to increase into the future [27]. The key drivers for this positive prospect of oxygenate usage are CO₂-emission legislation and the target of biofuel share in the transportation sector [28].

Fundamental combustion experimental studies on fully blended gasoline fuels are quite scarce. A few studies have focused on measuring their auto-ignition behavior in shock tubes and rapid compression machines. The work of Gauthier et al. [29] on the autoignition of full-blend, non-oxygenated, research grade gasoline (RD 387) in a shock tube has been extensively used to validate various surrogate models reported in the literature [30–32]. The shock tube experiments were conducted over relatively high temperatures of 850–1280 K, at pressures in the range 15–60 atm and at varying equivalence ratios of $\phi = 0.5$, 1.0, and 2.0. To study the auto-ignition of gasoline at low temperatures, Kukkadapu et al. [33] measured ignition delay data of RD 387 using an RCM in the temperature range 640–955 K, at compressed gas pressures of 20 and 40 bar for equivalence ratios ranging from 0.3 to 1.0. In this work, ignition delay data were simulated using a quaternary gasoline surrogate model developed by Mehl et al. [30], consisting of 57% *iso*-octane, 16% *n*-heptane, 23% toluene and 4% 2-pentene (by volume). Recently, Kukkadapu et al. [34] studied the autoignition characteristics of two gasoline surrogates, suggested by Mehl et al. [30] and Gauthier et al. [29], using an RCM at elevated pressure and low temperature conditions with lean fuel/air mixtures. Sarathy et al. [25] investigated the ignition characteristics of two non-oxygenated alkane-rich FACE (Fuels for Advanced Combustion Engines) gasoline fuels and their corresponding PRF blend in a shock tube and a rapid compression machine and proposed multi-component surrogates containing *iso*-pentane, 2-methylhexane, *iso*-octane, *n*-butane, *n*-heptane and toluene. In

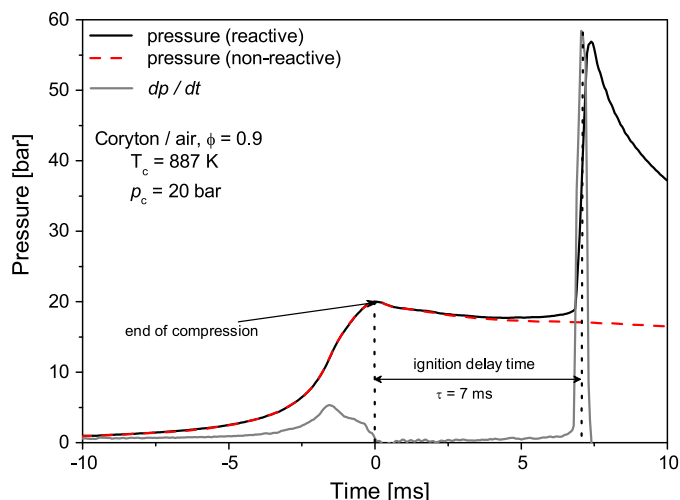


Fig. 2. Representative pressure profile and pressure derivative from rapid compression machine experiments. Definition of ignition delay time is shown.

another recent study, Sarathy et al. [35] examined the ignition behavior of two high-sensitivity FACE gasoline fuels and demonstrated the superior performance of multi-component surrogates over binary and ternary ones. To the authors' knowledge, detailed autoignition study of full-blend gasoline containing oxygenates such as ethanol is not available in the literature. The octane number variation due to the addition of ethanol to gasoline fuel surrogates such as PRF and TPRF have recently been investigated in engine studies [36,37]. A couple of studies [38,39] have been conducted to suggest surrogate formulations and chemical kinetic modeling of fuels containing ethanol.

The objectives of this study are twofold. First, we aim to investigate the autoignition characteristics of two oxygenated gasoline fuels over a wide range of pressure (10–40 bar), temperatures (650–1250 K) and equivalence ratios in a shock tube and a rapid compression machine. Second, we compare the ignition delay times of gasolines with ternary, quaternary and multi-component surrogates to suggest suitable surrogates for ethanol-containing gasolines. A chemical kinetic model is developed to simulate ignition delay times of the multi-component surrogate and to assess the fidelity of the surrogate model in capturing the auto-ignition behavior of gasoline.

2. Methodology

2.1. Shock tube

All measurements were made in a stainless steel shock tube of 63.5 mm inner diameter with a driver section 3 m in length, as described in detail previously [40]. The driver and driven sections of the shock tube are separated by a diaphragm chamber which houses two aluminum diaphragms, which are scored to a specific depth depending on the desired pressure behind the reflected shock wave. Shock waves are generated on pressure bursting of the diaphragms causing instantaneous heating of the fuel–air mixture contained in the 5.7 m long driven section. Shock velocities are determined using six piezoelectric pressure sensors (PCB 113A) mounted at 10, 150, 430, 710, 1025 and 2585 mm from the end-wall of the driven section. An additional pressure sensor (Kistler 603B) is mounted directly on the end-flange of the tube to detect the pressure rise due to ignition. The incident shock speed at the end-wall was determined using linear extrapolation of the axial velocity profile and the incident shock speed was applied to calculate the reflected temperature (T_5) and pressure (p_5) behind

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