



Rapid-compression machine studies on two-stage ignition characteristics of hydrocarbon autoignition and an investigation of new gasoline surrogates



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ABSTRACT

Iso-octane-based surrogates have been widely used to represent gasoline fuel combustion. The surrogate's autoignition characteristics have been of particular interest due to combustion issues associated with spark-ignition engine knocking or controlled autoignition. In this study, the autoignition of several hydrocarbons was measured and analyzed using a rapid compression machine. The two-stage ignition characteristics of gasoline and iso-octane were compared and the differences analyzed from the viewpoint of the effect of the fuels' chemical structure on low-temperature oxidation processes and internal isomerization. Finally, the reliability of iso-octane as a gasoline surrogate was examined, and other surrogates were suggested. Our results indicated that surrogates containing ~20% cyclic alkenes were better able to simulate gasoline autoignition, including two-stage ignition characteristics.

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1. Introduction

Iso-octane-based surrogates have long been used to simulate gasoline-fuel combustion [1–9]. It is mainly because the iso-octane's physical properties (e.g., density and boiling point) and the RON (Research Octane Number) is similar to that of gasoline. Accordingly, studies on iso-octane combustion processes have played a key role in providing a better understanding of the gasoline-fueled combustion mechanism, e.g., the spark-ignition (SI) engine. Among the combustion characteristics, autoignition has been investigated to identify the knocking phenomenon in gasoline engines and has recently been highlighted in gasoline homogeneous charge compression ignition (HCCI) or controlled autoignition (CAI) engine combustion [10–15].

The autoignition chemistry of alkanes was modeled by Hu et al. [16], and the low-temperature kinetics was established by Pilling et al. [17], Battin [18], Simmie [19], and Griffiths [20]. Also, Chun

et al. [21] successfully modified the autoignition process of Hu et al. [16] to predict SI engine knock. Dec et al. [22,23] and Ming et al. [24] analyzed gasoline HCCI combustion using iso-octane as a surrogate; Ming et al. [24] also suggested that the two-stage ignition characteristics of fuel are an important parameter in HCCI combustion.

Two-stage combustion refers to the high- and low-temperature regimes of autoignition characteristics, as determined by the negative temperature coefficient (NTC) shown in Fig. 1. Walker and Morley [25] describe the first stage of the two-stage ignition as being initiated by fuel oxidation via complex kinetics involving alkyl (R), alkylperoxy (RO₂), and peroxyhydroperoxyalkyl (O₂ROOH) radicals, in readily reversible addition and isomerization reactions. These processes can lead to degenerate chain branching, propagated predominantly by OH radicals, and are overall, modestly exothermic. The heat release raises the temperature of the reacting mixture. There are also competitive abstraction processes, which, due to the reversibility of the low-temperature branching sequences, tend to dominate the chemistry as the reactant temperature increases (typically to within the temperature range of 750–850 K). This yields alkenes (among other intermediate products) and HO₂ radicals, which tend to predominate in the chain propagation. Only a small amount of heat is released from

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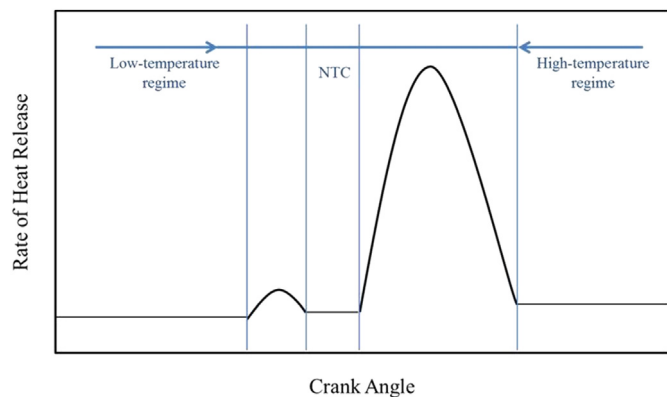


Fig. 1. Typical heat release curve from two-stage ignition. NTC corresponds to the negative temperature coefficient regime.

these reactions, following the first stage, as shown in Fig. 1. This kinetic transition from degenerate branching to a non-branching mechanism is the underlying cause in the shift of the overall reaction rate to negative temperature dependence. The HO_2 propagation constitutes an important route to hydrogen peroxide formation, the decomposition of which becomes more vigorous as the reactant temperature increases further, and initiates the second stage of ignition (Fig. 1). The heat release rate then becomes much more vigorous, raising the reactant temperature still further, eventually favoring chain-branching reactions involving O and OH radicals and small molecular intermediates, such as CO.

The research of Ming et al. [24] indicated that the octane number of the fuel determines the occurrence of two-stage ignition; specifically, for higher octane numbers (gasoline-like fuel), the fuel's autoignition reaction becomes suppressed at low temperatures. In this case, only a small amount of heat is released during the first stage, resulting in single-stage fuel ignition. Dec et al. [22,23] classified iso-octane, which has a high octane number similar to that of gasoline, into a single-stage ignition fuel for simulating gasoline HCCI combustion.

The octane number is a useful index for classifying the combustion characteristics of fuels; however, it provides only a rough empirical estimate. When considering two-stage ignition and various thermokinetic processes under nonisothermal conditions, it is necessary to examine the fuel's low-temperature oxidation processes. In the present study, iso-octane autoignition was compared with that of gasoline to verify its combustion characteristics; the comparison included two-stage ignition at low temperatures and high-pressure conditions. The two-stage ignition characteristics of several fuels were examined and compared with iso-octane to characterize the difference between first-stage and two-stage ignition. Based on these results, a new surrogate blend was suggested to better simulate gasoline's autoignition characteristics.

2. Experiments

2.1. The rapid compression machine (RCM)

A rapid compression machine (RCM) is an experimental apparatus used to simulate the single compression event of fuel-air mixture. The RCM uses pneumatic power to move the piston from bottom dead center (BDC) and hydraulic power to stop the entire piston assembly instantly at top dead center (TDC). Upon reaching TDC, the piston remains fixed at this location, and the volume of the cylinder is maintained at a constant value. In short,

the instant compression that occurs from BDC to TDC is used to simulate the compression stroke of an internal combustion engine. After the stroke, the RCM functions as a constant volume chamber. Thus, the RCM is a device by which, under the conditions of temperature and pressure at TDC (obtained from the compression stroke), combustion can be solely and quantitatively analyzed while excluding various external effects such as volume changes and fluid flow that may occur after compression.

The RCM used in this study was based on the design by Mittal [26] and modifications by Lee et al. [27]. Fig. 2 shows a schematic diagram of the RCM. The initial temperature and pressure could be set at 295–600 K and 1–3 bar, respectively, using the mixture preparation vessel. The maximum compression temperature and pressure are limited by durability of hardware. Considering the maximum temperature and pressure for each of seals and optical windows of the combustion chamber, the maximum temperature and pressure of RCM were set to 1100 K and 50 bar, respectively.

The pneumatic power was used to adjust the compression ratio and time over the ranges of 13–21 and 15–40 ms, respectively. Compression time of 15–40 ms is much longer than the ignition delay. Therefore, it would be possible for the mixture to react during compression. Therefore the result from the RCM should be analyzed carefully. However, in this study, it is important to have the comparison of relative combustion characteristics, and the subject of this study, premixed compression ignition is also influenced by the chemical reaction during the compression process. Therefore, it would be reasonable to assume that RCM could be appropriate in this study.

In applying the design of Mittal and Lee [26,27], the removal of the roll-up vortex was a key consideration. The roll-up vortex refers to fluid flow initiated by a temperature rise in the cylinder core during compression, while the wall temperature remains cold. An important feature of a RCM is that it can chemically analyze the combustion process after the compression stroke by excluding other external effects; thus, fluid flow must be minimized or eliminated for accurate measurement. To address this issue, in this study, we applied a creviced piston, which was shown by Mittal et al. [28] to eliminate the roll-up vortex.

2.2. Validation of the RCM

Prior to the main work, experiments were performed to test the reliability of the proposed RCM, and these were compared with results reported in the literature. Figs. 3 and 4 show the reproducibility test results. Under the initial conditions of 1 bar and 297 K, after the 5 times of test, it is showed that the average and standard deviation of the maximum pressure after the compression of inert gas were 27.5 bar and 0.22 bar, respectively, and the average and standard deviation of the ignition delay after the compression of PRF were 7 ms and 0.22 ms, respectively. Therefore, it would be possible to verify that its reproducibility is excellent.

Fig. 5 shows the comparison results from an ignition delay test of n-heptane combustion; likewise, the test results were consistent with those in the literature [29].

2.3. Experimental conditions

All experiments were conducted under initial temperature and pressure of 297 K and 1 atm, respectively. Two experiments were performed. The first experiment was designed to examine the autoignition characteristics of individual hydrocarbons, such as ignition delay, based on the pressure–time histories. The fuels and their properties are shown in Table 1. The second experiment was performed to identify potential gasoline surrogates other than iso-octanes. The main goal of the second experiment was to select a

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