



# First-stage ignition delay in the negative temperature coefficient behavior: Experiment and simulation



Peng Zhang<sup>a,b</sup>, Weiqi Ji<sup>a,b</sup>, Tanjin He<sup>a,c</sup>, Xin He<sup>a,c,\*</sup>, Zhi Wang<sup>c</sup>, Bin Yang<sup>a,b</sup>, Chung K. Law<sup>a,d</sup>

<sup>a</sup> Center for Combustion Energy, Tsinghua University, Beijing 100084, China

<sup>b</sup> Key Laboratory for Thermal Science and Power Engineering of MOE, Tsinghua University, Beijing 100084, China

<sup>c</sup> State Key Laboratory of Automotive Safety and Energy, Tsinghua University, Beijing 100084, China

<sup>d</sup> Department of Mechanical and Aerospace Engineering, Princeton University, Princeton, NJ 08544, USA

## ARTICLE INFO

### Article history:

Received 16 November 2015

Revised 2 March 2016

Accepted 2 March 2016

Available online 22 March 2016

### Keywords:

First-stage ignition

Negative temperature coefficient

Rapid compression machine

Low-temperature mechanism

## ABSTRACT

The existence of the first-stage ignition delay (FID) negative temperature coefficient (NTC) behavior was confirmed by rapid compression machine experiments using iso-octane and methyl-cyclohexane. The first-stage NTC behavior of iso-octane is observed in the temperature range 757–782 K under 20 bar and  $\phi = 1$ . For methyl-cyclohexane, the observed first-stage NTC temperature range is 750–785 K under 15 bar and  $\phi = 0.5$ . In further iso-octane experiments, the FID is found to be sensitive to the  $O_2$  concentration and insensitive to the dilution gas and fuel concentrations. The effects of the FID and its NTC behavior on the total ignition delay NTC were analyzed using a detailed *n*-heptane mechanism. The contributions to the total ignition delay NTC from the reduced second-stage initial temperature, pressure, and less reactive species pool, together with the NTC of FID were discussed quantitatively. For the first-stage NTC behavior, five competing reactions were identified as being important based on sensitivity analysis, reaction pathway analysis, and simplified mechanism method. They are the backward reaction of second  $O_2$  addition,  $RO_2 \rightleftharpoons \text{alkene} + HO_2$ ,  $QOOH \rightleftharpoons \text{cyclic-ether} + OH$ ,  $QOOH \rightleftharpoons \text{alkene} + HO_2$ , and the beta scission reaction of the alkyl radical. Their competition with the low-temperature branching channel finally leads to the first-stage NTC behavior.

© 2016 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

## 1. Introduction

Under practical engine conditions, the ignition characteristics of hydrocarbon fuels can be divided into two classes: those with single-stage auto-ignition such as aromatics and alcohols, and those with two-stage auto-ignition such as *n*-paraffins, iso-paraffins, and cyclo-paraffins, with the latter usually observed at temperatures below 850 K. When two-stage ignition occurs, the first-stage ignition assumes an essential role because the second-stage ignition depends on the heat release and intermediate species generated from it. Furthermore, the negative temperature coefficient (NTC) regime of the total ignition delay spans exactly around this temperature range, which is relevant to engine knock and other related combustion phenomena [1,2]. Since hydrocarbons with two-stage ignition typically constitute more than 50% of practical fuels [3], engine processes controlled by combustion kinetics such as homogeneous charge compression ignition (HCCI) would occur in two stages as well: a low-temperature heat release

(LTHR) stage followed by a high-temperature heat release (HTHR) stage. It is then significant to note that fuels with two-stage ignition have been found to offer significant advantages in controlling combustion phasing and extending the HCCI operation range [4,5]. Consequently, it is essential to better understand the phenomena associated with the first-stage ignition delay (FID) so that better control strategies can be developed for optimal fuel economy and lower pollutant emissions.

Extensive studies have been conducted on the first-stage ignition and cool flames. Specifically, Minetti et al. [6] conducted a series of *n*-heptane experiments using a Rapid Compression Machine (RCM), and found that the amplitude of the first-stage heat release decreases sharply with temperature, and that the delay does not depend on the rate of compression, which in turn implies that no significant reactions occur during compression. Fieweger et al. [7] investigated the auto-ignition of *n*-heptane and iso-octane mixtures in a shock tube under the condition of  $p = 40$  bar and  $\phi = 1$ , with the first-stage ignition detected by CH emission instead of pressure history. Results show that the FID of the mixture is within the range limits of the pure fuels. Herzler et al. [8] studied the ignition behavior of lean *n*-heptane/air mixtures at low to intermediate temperatures and high pressures in a shock tube, with the pressure as high as 50 bar and equivalence

\* Corresponding author at: Tsinghua University, Center for Combustion Energy and State Key Laboratory of Automotive Safety and Energy, Room 112, Automobile Research Institute, Tsinghua University, Beijing 100084, China. Fax: +861062798267.

E-mail address: [hexin1976@tsinghua.edu.cn](mailto:hexin1976@tsinghua.edu.cn), [hexin.tsinghua@gmail.com](mailto:hexin.tsinghua@gmail.com) (X. He).

ratio varying from 0.1 to 0.4. Two-stage ignition was observed in the temperature range 720–850 K, with the FID decreasing from 2 ms to 0.1 ms with increasing temperature, and is independent of equivalence ratio. Vanhove et al. [9] studied low-temperature auto-ignition of five binary hydrocarbon mixtures at undiluted stoichiometric conditions using an RCM, and found that the FID decreases with increasing temperature for all the mixtures studied. Kuwahara et al. [4,10] studied the effect of fuel, O<sub>2</sub>, and N<sub>2</sub> concentrations on FID using a detailed *n*-heptane mechanism generated by KUCRS (knowledge-basing utilities for complex reaction systems) [11], which is an automatic mechanism generation code. Under the condition of  $\phi=0.5$ ,  $p=1$ ,  $T=759$  K, the FID was found largely depending on the O<sub>2</sub> concentration and is insensitive to the fuel and N<sub>2</sub> concentrations.

It is further noted that while most studies of the two-stage ignition and NTC behavior were conducted for homogeneous systems, recent investigations have extended the study to inhomogeneous, diffusive-convective systems, notably those of the non-premixed counterflow [12–14]. A secondary S-curve was found on the lower branch of the conventional S-curve, signifying the feasibility of NTC-controlled ignition in diffusive, nonpremixed media. The results further indicate that the FID may possess a minimum value, which is confirmed by the simulation study using the detailed mechanism. More recently, the first-stage NTC behavior of *n*-heptane was observed by Campbell et al. [15]. Ignition delay times of *n*-heptane were measured at 6.1–7.4 atm and 651–823 K using constrained reaction volume approach facilitated with staged-driver gas filling strategy, driver section extensions, driver inserts, and driver gas tailoring. The first-stage NTC behavior was clearly observed. Cyclo-paraffin may also exhibit the first-stage NTC phenomenon. Mittal and Sung [16] investigated auto-ignition of methyl-cyclohexane (MCH) at the elevated pressure of 25.5 bar at  $\phi=1.0$  and 15.1 bar at  $\phi=0.5$ –1.5. It was found that the FID first decreases with increasing temperature, levels off, and then increases slightly. This behavior was suggested to be caused by the combined influence of the heat loss and the definition of the FID on the basis of inflection point during the first-stage reactivity [16]. Although higher initial temperature results in shorter induction time, more time is needed for the first-stage ignition to overcome the pressure drop due to heat loss to exhibit a net pressure rise and an apparent inflection point, leading to a prolonged measured FID. Battin-Leclerc [17] cited RCM experimental data from [18], showing that the FID of cyclohexane possesses an NTC regime in 770–800 K under the condition of 7–9 bar and  $\phi=1$ , although this interesting phenomenon was not explained in the original paper of the experiments [18]. We finally note that there has been no data reported on the first-stage NTC behavior of iso-paraffins.

Considering the importance of two-stage ignition and limited experimental data on the first-stage NTC behavior, the present study aimed to investigate such a behavior with the use of *n*-heptane, iso-octane, and methyl-cyclohexane. The investigation involves first experimentally demonstrating the first-stage NTC behavior, substantiated by effects of O<sub>2</sub>, dilution gas, and fuel concentration on FID. Then the effects of the first-stage NTC behavior on the total ignition NTC behavior are demonstrated using a detailed *n*-heptane mechanism. Finally sensitivity analysis, reaction pathway analysis, and simplified mechanism analysis results are shown to identify five important reactions for the first-stage NTC behavior.

## 2. Experiment

### 2.1. The rapid compression machine

All experiments were conducted in the well-controlled and well-characterized environment of the RCM developed at Tsinghua

University. It is a typical pneumatic-driven, hydraulic-deceleration RCM. Crevice piston design is applied and the dimension of the crevice (provided in the Supplemental Material) is optimized to minimize the effect of the roll-up vortex. The premixed mixture charged in the combustion chamber is compressed to the high pressure, high temperature condition in 25–30 ms. Half of the pressure and temperature rise occurs within 3 ms before the end of compression, such that effects from potential reactions during compression are minimized. The stroke of the piston is 0.5 m, providing a large clearance height to minimize the heat loss. A piezoelectric pressure transducer (Kistler 6125C) is mounted on the combustion chamber to measure the pressure. The temperature at the end of compression is calculated using the adiabatic core assumption. The ignition delay is determined by the pressure history. Detailed descriptions of the RCM are given in previous papers [19,20].

As discussed in our previous work [19], the uncertainty of ignition delay mainly comes from three parts, namely the uncertainties of the measured pressure, measured temperature, and the compression process. The pressure part primarily originates from the accuracy of the initial pressure, the pressure transducer in the combustion chamber, and the charge amplifier. The temperature part mainly consists of the uncertainties of the initial temperature and pressure transducer (used to deduce temperature after compression). The uncertainties due to compression is calculated based on the Livengood–Wu integral from the start to the end of compression. The combined uncertainty is the square root of the sum of squares of the three parts. All the experimental data used in this paper and the corresponding estimated uncertainties are provided in the Supplemental Material.

### 2.2. Test mixtures

The test mixtures were prepared using a stainless steel mixing tank. The tank was evacuated to 0.1 Torr before the fuel injection. The partial pressure of the fuel was kept less than 50% of its vapor pressure to avoid potential condensation. The vapor pressure of *n*-heptane, iso-octane, and methyl-cyclohexane are high enough so that all experiments reported herein were conducted under room temperature. The mixture composition was determined by the partial pressure of the mixture components. The temperature after compression was varied by adjusting the compression ratio.

Three pure fuels, i.e. *n*-heptane, iso-octane, and methyl-cyclohexane, were investigated as they represent normal, branched, and cyclo-alkanes, respectively. Table 1 shows the mixture composition used in this study. Experimental results with mixtures #X1-2 and #Y1-2 have been reported in our previous paper investigating the buffer gas effect [19]. These ignition delay results are not listed here, but the data were analyzed further in view of the FID to maintain consistency. Experimental results with mixtures #A1, #A2, #B1, and #B2 are presented first. It is noted that mixtures #A1 and #B1 are combustible mixtures, while mixtures #A2 and #B2 have similar composition as those of #A1 and #B1, except O<sub>2</sub> was substituted by N<sub>2</sub>. Since the specific heat ratio of N<sub>2</sub> is similar to that of O<sub>2</sub>, mixtures #A2 and #B2 were used to characterize the heat loss after compression. For example, experiments using #A1 and #A2 with the same initial condition result in the same pressure profile before heat release from reactions. So the difference between the pressure profiles of #A1 and #A2 is the net pressure rise corresponding to the heat release of reactions.

To study the NTC behavior of the FID, two factors should be considered when choosing the mixture composition and experimental conditions. First, the  $\tau_{1,\min}$  (the shortest FID) should be long enough to be observed by RCM, recognizing that it is very difficult to identify the pressure inflection point if the ignition delay is shorter than 5 ms. Short ignition delay could also cause large

Download English Version:

<https://daneshyari.com/en/article/6594079>

Download Persian Version:

<https://daneshyari.com/article/6594079>

[Daneshyari.com](https://daneshyari.com)