

Fuel Design Concept for Robust Ignition in HCCI Engine and Its Application to Optimize Methane-Based Blend

Hiroki Tanaka, Shunsuke Somezawa, and Takahiro Sako
Osaka Gas Co., Ltd.

Yasuyuki Sakai and Hiromitsu Ando
University of Fukui

Kazunari Kuwahara
Osaka Institute of Technology

ABSTRACT

A fuel design concept for an HCCI engine based on chemical kinetics to optimize the heat release profile and achieve robust ignition was proposed, and applied to the design of the optimal methane-based blend.

Ignition process chemistry of each single-component of natural gas, methane, ethane, propane, *n*-butane and isobutane, was analyzed using detailed chemical kinetic computations. Ethane exhibits low ignitability, close to that of methane, when the initial temperature is below 800 K, but higher ignitability, close to those of propane, *n*-butane and isobutane, when the initial temperature is above 1100 K. Furthermore, ethane shows a higher heat release rate during the late stage of the ignition process. If the early stage of an ignition process takes place during the compression stroke, this kind of heat release profile is desirable in an HCCI engine to reduce cycle-to-cycle variation during the expansion stroke.

According to results from engine operation tests using dual-component fuels with methane as the primary component and ethane, propane, *n*-butane and isobutane as the secondary component, methane/ethane shows a lower COV of IMEP when CA50 is set at the same timing for the expansion stroke. Furthermore, methane/ethane also shows a lower knocking intensity when CA50 is set at the same timing, close to the knocking limit, due to its lower in-cylinder pressure rise rate.

These results suggest that methane/ethane can be the optimal methane-based blend for an HCCI engine to achieve both better fuel economy and higher performance by its robust ignition and high anti-knocking properties.

CITATION: Tanaka, H., Somezawa, S., Sako, T., Sakai, Y. et al., "Fuel Design Concept for Robust Ignition in HCCI Engine and Its Application to Optimize Methane-Based Blend," *SAE Int. J. Engines* 7(2):2014, doi:10.4271/2014-01-1286.

INTRODUCTION

The potential for homogenous charge compression ignition (HCCI) engines using a variety of fuels has been examined with a view to achieving higher thermal efficiency and lower NO_x emissions [1, 2, 3, 4, 5, 6]. Sako *et al.*, have considered the requirements for HCCI engines using natural gas, a fuel with extremely low ignitability, and established that the operating range under high-load conditions is limited by rapid combustion with heavy knocking and cycle-to-cycle variation [7, 8]. This narrow operating range is the main obstacle for an HCCI engine. Many studies have examined ways to expand the operating range, such as supercharging, exhaust gas recirculation (EGR), and stratification [9, 10, 11, 12, 13, 14].

Kuwahara *et al.*, have analyzed the ignition process chemistry of *n*-heptane using a detailed chemical kinetic model and a contribution matrix method. They have also proposed an approach that divides the hydrocarbon ignition process into five phases and seven reaction groups listed below [15, 16, 17, 18]. The thermal ignition preparation phase is the process in which the relatively slow heat release by the H₂O₂ loop reaction raise the temperature to the thermal ignition point at which the branching chain reactions of the hydrogen oxygen system become dominant.

- Phases: LTO preparation / LTO (low-temperature oxidation) / NTC (negative temperature coefficient) / Thermal ignition preparation / Thermal ignition
- Reaction groups: Fuel series chemistry / Fuel fragment

chemistry / H₂O₂ loop chemistry / HO₂ chemistry / Hydrogen oxygen system chemistry / CO₂ chemistry / HACA (H abstraction C₂H₂ addition) chemistry

Because HCCI combustion is controlled by chemical reactions without the help of forced ignition such as spark discharge or diesel injection, the ignition robustness against in-cylinder disturbances during the expansion stroke, is low and cycle-to-cycle variation occurs easily. This is because slow heat release during the thermal ignition preparation phase is less than the decrease in the internal energy due to expansion.

This paper presents an analysis based on chemical kinetics of the ignition process chemistry of each component of natural gas, methane (CH₄), ethane (C₂H₆), propane (C₃H₈), *n*-butane (*n*-C₄H₁₀) and isobutane (*i*-C₄H₁₀), and proposes a fuel design concept which optimizes the composition of methane-based blends with a view to reducing cycle-to-cycle variation and expanding operating range of HCCI engines. Ignition robustness against in-cylinder disturbances during the expansion stroke for a dual-component methane-based blend with a primary component of CH₄ and a secondary component of C₂H₆, C₃H₈, *n*-C₄H₁₀ or *i*-C₄H₁₀ was estimated using detailed chemical kinetic computations, and the validity of the concept was verified through engine tests.

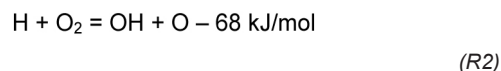
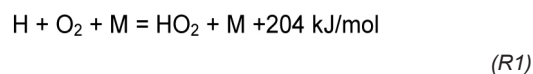
CHEMICAL KINETIC COMPUTATIONS UNDER ADIABATIC CLOSED CONSTANT-VOLUME CONDITIONS

Computation Method

A detailed chemical kinetic model for the oxidation of CH₄, C₂H₆, C₃H₈, *n*-C₄H₁₀, *i*-C₄H₁₀ and blends thereof was created using KUCRS. The model is composed of 145 chemical species and 658 elementary reactions. KUCRS (Knowledge-basing Utilities for Complex Reaction System) is the automatic detailed chemical kinetic model generation software, developed by Miyoshi [19, 20, 21, 22, 23]. The model includes H₂/O₂ reactions in the Leeds CH₄ model [24], and C₁ and C₂ reactions in the LLNL isooctane model [25]. The unimolecular reactions of alkyl peroxy radicals (ROO), hydroperoxy alkyl radicals (QOOH), and hydroperoxy alkyl peroxy radicals (OOQOOH) were discussed [22], and the molecular-size dependent fall-off rate constants of O₂ addition to alkyl radicals and thermal decomposition of hydroperoxy carbonyl compounds (HOOPO) were introduced [23]. Thermodynamic data were generated by THERM [21]. The computational ignition delay times of *n*-heptane and isooctane (2,2,4-trimethylpentane) in air using the model of primary reference fuels, the blends of *n*-heptane and isooctane, agree well with experimental data in a shock tube [18, 26].

Using the chemical kinetic computation software CHEMKIN-PRO, zero-dimensional calculations for the ignition of a mixture of a single-component fuel (CH₄, C₂H₆, C₃H₈, *n*-C₄H₁₀ or *i*-C₄H₁₀) and air (O₂: 21 vol. %, N₂: 79 vol. %) in an adiabatic closed constant-volume vessel were carried out. The initial fuel

molar concentration in each single-component fuel/air mixture was set at the equivalent of a calorific value of 32.20 MJ/m³, the initial O₂ molar concentration at 160.5 mol/m³, and the initial N₂ molar concentration at 603.7 mol/m³. These values correspond to the initial component molar concentrations in a CH₄/air mixture at an equivalence ratio of 0.5, initial temperature of 1000 K and initial pressure of 6.687 MPa (see Table 1.). The calorific value of the fuel was defined as the heat of formation that assumes complete oxidation. For each mixture, initial temperature was varied, while the initial fuel, O₂, and N₂ concentrations were kept constant to determine the relationship between initial temperature and ignition delay time. The point at which the rate of reaction (R2) exceeds that of reaction (R1) was defined as the thermal ignition point, and ignition delay time was determined [18].



Calculations were also carried out for a dual-component blended fuel with CH₄ as the primary component and C₂H₆, C₃H₈, *n*-C₄H₁₀ or *i*-C₄H₁₀ as its secondary component. First, the initial fuel, O₂ and N₂ molar concentrations for a mixture of air and natural gas (CH₄: 88 vol. %, C₂H₆: 6 vol. %, C₃H₈: 4 vol. %, *n*-C₄H₁₀: 1 vol. %, *i*-C₄H₁₀: 1 vol. %) were set to the equivalent of a calorific value of 32.20 MJ/m³, 160.5 mol/m³ and 603.7 mol/m³. Next, the initial CH₄, O₂ and N₂ molar concentrations for each dual-component fuel/air mixture were set to the equivalent of a calorific value of 24.54 MJ/m³, 160.5 mol/m³ and 603.7 mol/m³, which are the same values as those for the natural gas/air mixture. Then, the initial secondary fuel molar concentration was set to the equivalent of a calorific value of 7.659 MJ/m³, where the overall calorific value, made up of the calorific value of the CH₄ and that of the secondary fuel, was 32.20 MJ/m³ (see Table 2.).

Table 1. Computational conditions for single-component fuels in an adiabatic closed constant-volume vessel

Kinetics scheme	Detailed reaction model generated by KUCRS CH ₄ , C ₂ H ₆ , C ₃ H ₈ , <i>n</i> -C ₄ H ₁₀ , and <i>i</i> -C ₄ H ₁₀ 147 species, 658 elementary reactions
Solver	CHEMKIN-PRO
Reactor	0 dimensional, Adiabatic closed constant-volume
Reactants	Single-component fuel (CH ₄ , C ₂ H ₆ , C ₃ H ₈ , <i>n</i> -C ₄ H ₁₀ , or <i>i</i> -C ₄ H ₁₀)
Oxidant	Air (O ₂ : 21%, N ₂ : 79%)
Initial compositions	O ₂ : 160.5 mol/m ³ , N ₂ : 603.7 mol/m ³ , Fuel: 32.20 MJ/m ³ Equivalent to those in CH ₄ /air mixture at φ = 0.5, T ₀ = 1000 K, p ₀ = 6.687 MPa
Initial temperature	700 to 1200 K

Table 2. Computational conditions for dual-component blended fuels in an adiabatic closed constant-volume vessel

Kinetics scheme	Detailed reaction model generated by KUCRS CH ₄ , C ₂ H ₆ , C ₃ H ₈ , n-C ₄ H ₁₀ , and i-C ₄ H ₁₀ 147 species, 658 elementary reactions
Solver	CHEMKIN-PRO
Reactor	0 dimensional, Adiabatic closed constant-volume
Reactants	Dual-component fuel (CH ₄ /C ₂ H ₆ , CH ₄ /C ₃ H ₈ , CH ₄ /n-C ₄ H ₁₀ , or CH ₄ /i-C ₄ H ₁₀ .) Natural gas (CH ₄ : 88%, C ₂ H ₆ : 6%, C ₃ H ₈ : 4%, n-C ₄ H ₁₀ : 1%, i-C ₄ H ₁₀ : 1%)
Oxidant	Air(O ₂ :21%, N ₂ :79%)
Initial compositions	O ₂ : 160.5 mol/m ³ , N ₂ : 603.7 mol/m ³ , Fuel: 32.20 MJ/m ³ (CH ₄ : 25.54 MJ/m ³ , secondary fuel: 7.659 MJ/m ³)
Initial temperature	700 to 1200 K

Ignition Characteristics of Single-Component Fuels

Figure 1 shows the relationship between initial temperature and ignition delay time for a single-component fuel/air mixture. The ignition delay time is the longest for CH₄, followed in order by C₂H₆, C₃H₈, i-C₄H₁₀ and n-C₄H₁₀, in line with the generally known trend. However, the ignition delay time of C₂H₆ is as long as that of CH₄ when the initial temperature is low, approximately 800 K, and approaches those of C₃H₈, n-C₄H₁₀ and i-C₄H₁₀ when the initial temperature is high, approximately 1200 K. The ignition delay time of C₂H₆ is the shortest with initial temperature above about 1250 K. The apparent activation energy of C₂H₆ is higher than those of the other single-component fuels over a wide range of initial temperature, which has been reported from experimental studies using a flow reactor by Beerer *et al.* [27] and using a rapid compression machine by Furutani *et al.* [28].

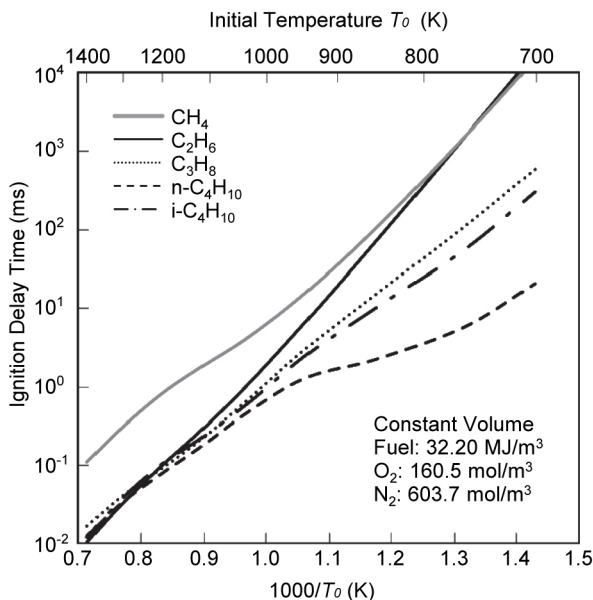


Figure 1. Relationship between initial temperature and ignition delay time for single-component fuels

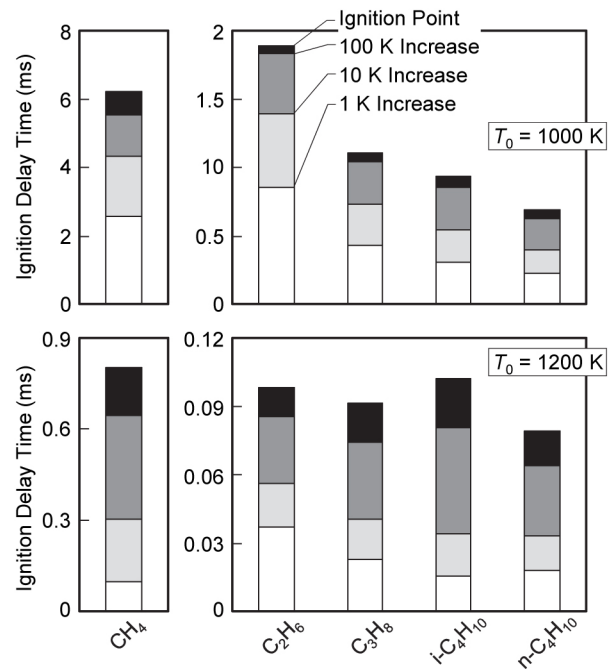


Figure 2. Ignition delay times and breakdowns of ignition delay time for single-component fuels

Figure 2 shows ignition delay times and breakdowns for initial temperatures of 1000 K and 1200 K. Under these conditions, the ignition process for each fuel begins in the thermal ignition preparation phase, bypassing LTO. Ignition delay time is divided at the points where temperature increases by 1 K, 10 K and 100 K. When the initial temperature increases from 1000 K to 1200 K, the ignition delay time of C₂H₆ approaches those of C₃H₈, n-C₄H₁₀ and i-C₄H₁₀, in particular because the ratio of the time between the start of the reaction and the point at which temperature increases by 10 K is shortened (see the white and light grey areas). When the initial temperature is 1200 K, the time between the points where temperature increases by 100 K and the ignition point is shorter for C₂H₆ than for C₃H₈, n-C₄H₁₀ and i-C₄H₁₀ (see the black areas in the lower graph).

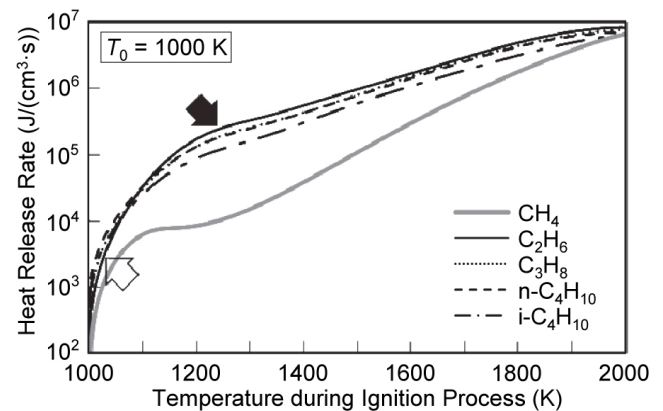


Figure 3. Profile of heat release rate for single-component fuels

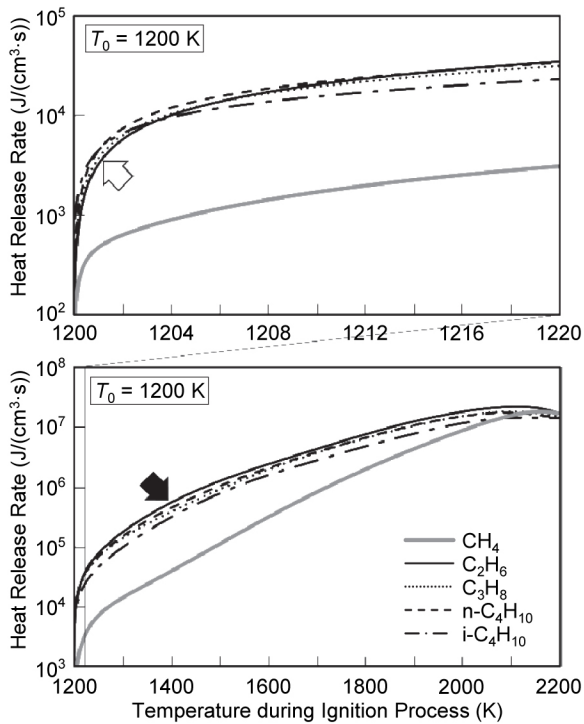


Figure 3. (cont.) Profile of heat release rate for single-component fuels

Figure 3 shows heat release rate plotted against increasing temperature during an ignition process. The heat release rate of C_2H_6 is lower than those of C_3H_8 , $n-C_4H_{10}$ or $i-C_4H_{10}$ when the temperature increase is in the region of several tens of K for an initial temperature of 1000 K, and when the temperature increase is several K for an initial temperature of 1200 K (see the white arrows). However, it is higher in most of the temperature range thereafter (see the black arrows).

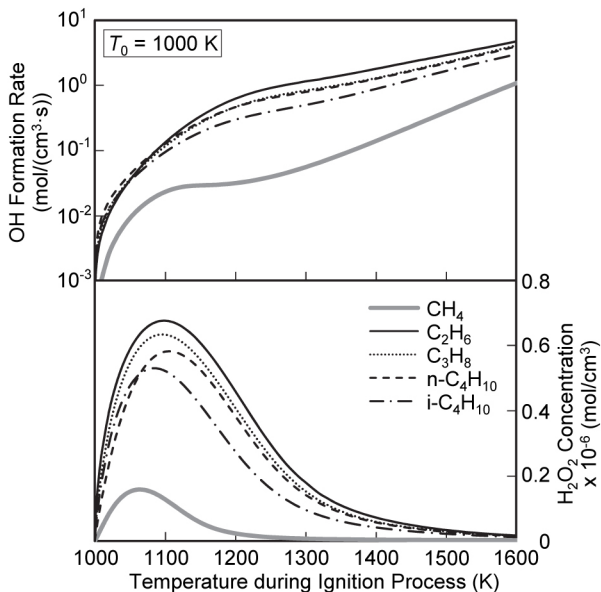
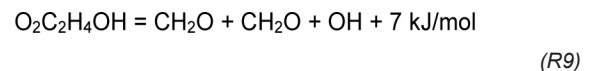
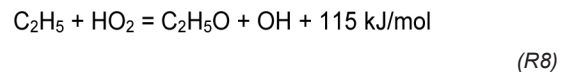
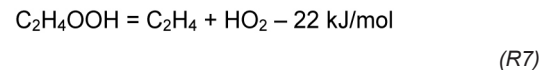
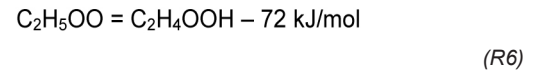
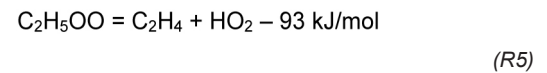
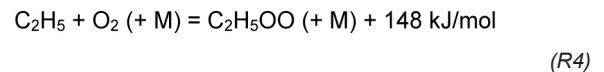
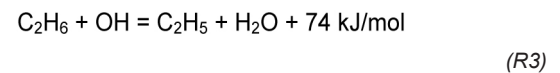


Figure 4. Profile of OH formation rate and H_2O_2 concentration for single-component fuels

The above heat release profiles for C_2H_6 can be explained as follows. Figure 4 shows overall OH formation rate (sum of OH formation rates of all related forward reactions) and H_2O_2 concentration against increasing temperature during an ignition process, for an initial temperature of 1000 K. According to a reaction path analysis using the contribution matrix method proposed by Kuwahara *et al.* [15, 16, 17, 18], the major reaction paths in the fuel series chemistry of C_2H_6 are reactions (R3), (R4), (R5), (R6), (R7). In other words, the HO_2 formation path is dominant and the OH formation paths (R8) and (R9) are not significant. Furthermore, H_2O_2 accumulates from the HO_2 removal reaction, $HO_2 + HO_2 \rightarrow H_2O_2 + O_2$. Within the lower temperature range where fuel series chemistry is dominant, OH formation and heat release are both slow, but in the higher temperature range, once the H_2O_2 loop [15] becomes active, heat release is accelerated by the high H_2O_2 concentration.



Ignition Characteristics of Dual-Component Fuels

Figure 5 shows the relationship between initial temperature and ignition delay time for a dual-component fuel/air mixture. The ignition characteristics of dual-component fuels reflect those of the secondary fuels, *i.e.*, the ignition delay time is the longest for CH_4 , followed in order by CH_4/C_2H_6 , CH_4/C_3H_8 , $CH_4/i-C_4H_{10}$ and $CH_4/n-C_4H_{10}$. The ignition delay time of CH_4/C_2H_6 is as long as that of CH_4 when initial temperature is low and approaches that of the other dual-component fuels as initial temperature increases. The proportion of C_2H_6 in natural gas is greater than that of C_3H_8 , but the relationship between initial temperature and ignition delay time for natural gas is close to that for CH_4/C_3H_8 .