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Study on combustion and ignition characteristics of ethylene, propylene, 1-butene and 1-pentene in a micro flow reactor with a controlled temperature profile

Shogo Kikui [a,*], Hisashi Nakamura [a], Takuya Tezuka [a], Susumu Hasegawa [a], Kaoru Maruta [a,b]

[a] Institute of Fluid Science, Tohoku University, 2-1-1 Katahira, Aoba-ku, Sendai, Miyagi 980-8577, Japan
[b] ICE Lab., Far Eastern Federal University, 8, Sukhanova Str., Vladivostok 690950, Russia

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ABSTRACT

Weak flames of four alkenes (ethylene, propylene, 1-butene and 1-pentene) were observed using a micro flow reactor with a controlled temperature profile to investigate their combustion and ignition characteristics. Weak-flame based investigation which enables to elucidate general ignition property of each fuel was conducted. One-dimensional computations with detailed reaction mechanisms were conducted to compare and analyze experimental results. Single luminous zone of hot flame was observed for all four alkenes. The order of weak flame position was ethylene, 1-pentene, 1-butene and propylene from the lower temperature side and thus the reactivity of those fuels are higher in the same order. Computational results captured the order of the experimental weak flame position. Alkene results were compared with alkane results in terms of carbon number and the order of weak flame position agreed between the alkene and alkane results. Weak flame structures of alkenes were analyzed using the computations. A similar overall flame structure was obtained for the four alkenes from the mole fraction profiles of major species. Rate of consumption was investigated to clarify fuel consumption of alkenes. Reactions with O, H and OH radicals proceed in the fuel consumption of alkenes. H-atom addition reaction of alkene where the reaction occurs at the double bond of the alkene is a unique reaction compared with the alkane and consumes a significant amount of the fuel. H-atom abstraction reaction with OH, which is important in alkane, is also important in the alkene consumption. Reaction path analysis was conducted to examine the high reactivity of ethylene. OH production through HO_2 in the initial stage of oxidation is important against the weak flame position. Ethylene has a high rate of HO_2 production compared with the other alkenes through $HCO + O_2 <=> CO + HO_2$, $C_2H_5 + O_2 <=> C_2H_4 + HO_2$ and $C_2H_3 + O_2 <=> C_2H_2 + HO_2$, which results in a high reactivity.

1. Introduction

Combustion characteristics of alkenes are receiving interest not only because they are essential components in practical fuels but they are also a key intermediate in alkane oxidation [1,2]. Alkanes are main components of practical fuels and understanding their reaction process in detail is critical to improve various combustion devices. Many reaction mechanisms were developed for alkanes but further improvement is required. As a key intermediate in alkane oxidation, understanding alkene reaction process could definitely lead to the improvement of understanding alkane. Despite the importance of understanding alkenes, there are limited numbers of researches on them compared with the extensively-studied alkanes. Therefore, alkene combustion is not as clearly understood as alkanes. Ethylene is mostly studied among all alkenes since it is also known as an important intermediate for soot formation. Modeling of ethylene combustion was done in studies such as in Xu and Konnov [3] and Carrier et al. [4]. Ignition characteristics were studied experimentally with shock tube and rapid compression machines by Sexena et al. [5] and Kumar et al. [6]. Larger alkenes like pentene have been broadly studied since it is seen in gasoline oxidation. Kukkadapu et al. [7] added pentene to a gasoline surrogate in order to study its effect on ignition delay. Prabhu et al. [8] studied pentene oxidation and its interaction with nitric oxide. Experimental and modeling studies of pentene were done by Minetti et al. [9], Ribaucour [10] and Touchard et al. [11,12]. Studies on lower alkenes, such as propylene [13–15] and butene [16], or studies focusing on alkene isomers [17–19] were also seen in the study of alkenes. However, not many researches apply multiple alkenes with different carbon size and compare each other. Most of the comparisons for alkenes were done with their conjugate alkanes or their own isomers.

* Corresponding author. Fax: +81 22 217 5296.
E-mail address: kikui@edyn.ifs.tohoku.ac.jp, hcrxxxii@gmail.com (S. Kikui).

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Nomenclature

A	cross-sectional area [cm^2]
A_i	frequency factor of elementary reaction [reaction-dependent]
c	specific heat [J/(g K)]
d	inner diameter of reactor [mm]
h	specific enthalpy [J/g]
j	mass flux [g/(cm^3 s)]
K	total number of species [–]
M	mass flow rate [g/s]
Nu	Nusselt number [–]
p	pressure [atm]
T	temperature [K]
V_k	diffusion velocity [cm/s]
u	mean flow velocity [cm/s]
W	molecular weight [g/mol]
x	spatial coordinate [mm]
X	mole fraction [–]
Y	mass fraction [–]
[A]	mole density of species A [mol/cm^3]
λ	thermal conductivity [W/(cm K)]
ρ	density [g/cm^3]
$\dot{\omega}$	molar production rate [mol/(cm^3 s)]
t	camera exposure time [s]

Subscripts

ad	adiabatic
i	elementary reaction index
k	species index
p	constant pressure
w	wall
0	origin

In this study, four alkenes of ethylene, propylene, 1-butene and 1-pentene were chosen as test fuels. These alkenes have different carbon number (C2–C5) with a similar molecular structure which is a straight chained alkene with a double bond locating at the end of the carbon chain. Therefore, carbon number dependence could be examined along with their individual combustion characteristics. In addition, the effect of double bond existence could be observed by the comparison against their conjugate alkanes of ethane, propane, *n*-butane and *n*-pentane.

To investigate combustion characteristics of the fuels, a micro flow reactor with a controlled temperature profile (denoted as micro flow reactor from here on) [20,21] was applied. The micro flow reactor is a recently introduced setup which could investigate combustion and ignition characteristics of given fuels in a well defined simple system. In particular, weak flame phenomenon is specific to this reactor and the weak-flame based investigation enables to elucidate general ignition property of given fuels [22–24]. A quartz tube with an inner diameter smaller than the quenching diameter of the introduced gas mixture is used as a reactor. A part of the reactor channel is heated using an external heat source to form a stable temperature gradient along the flow direction at the inner wall. The premixed fuel/oxidizer is introduced into the reactor and the various flames are observed. In the micro flow reactor, the pressure is constant the flow condition is laminar and the gas-phase temperature strongly depends on the wall temperature profile. In the past studies using the micro flow reactor [22–26], three types of flame were observed depending on inlet flow velocities: stable flat flames were observed at high flow velocities; unstable flames called *flames with repetitive extinction and ignition* (FREI) were observed at intermediate flow velocities; and stable flames with weak luminescence (weak flame) were observed at very low flow velocities. Special attentions has been given to the weak flame which is a unique phenomenon observed only in the micro flow reactor. Minaev et al. [27] showed that the weak flame branch in the micro flow reactor is on the ignition branch of the Fendell curve based on theoretical analysis. Oshibe et al. [26] applied dimethyl ether (DME) and Yamamoto et al. [22] applied *n*-heptane to the micro flow reactor and observed multiple weak flames both experimentally and numerically. In these studies, gas sampling and analysis indicated that the three-stage oxidation (three weak flames) corresponds to the typical cool, blue and hot flames known in the ignition phenomenon in other systems where cool flame is characterized by formation of CH_2O [28], blue flame is characterized by CH_2O consumption and CO and CH_4 formation [29], and hot flame is characterized by CO and CH_4 consumption and CO_2 formation [29]. In the weak flame region, gas-phase temperature profile nearly equals to the wall temperature profile and the rapid increase in the gas-phase is suppressed by the wall temperature. Therefore, the oxidation process is separated into different temperature ranges which are observed as multiple weak flames. The ignition process known to be a transient and spontaneous phenomenon is reproduced as a stationary phenomenon represented by steady weak flames in the micro flow reactor. Past studies by Hori et al. [23] and Suzuki et al. [24] focused on weak flame positions to examine reactivity of fuels. In these studies, gasoline primary referenced fuels (PRF) and diesel surrogate fuels were applied to the micro flow reactor. As the octane/cetane number increased/decreased the weak flame positions located at the higher temperature side which indicates lower reactivity. Another study by Hori et al. [30] investigated the effect of toluene addition to *n*-heptane and observed the inhibition of oxidation with toluene addition. The capabilities of the micro flow reactor to investigate general combustion and ignition characteristics of fuels have been demonstrated from these past studies.

The objective of this study is to investigate combustion and ignition characteristics of four alkenes (ethylene, propylene, 1-butene and 1-pentene) based on their weak flames using the micro flow reactor. Weak flames of these fuels were observed and analyzed using one-dimensional computation with detailed reaction mechanism. Weak flame position was used to compare reactivity of these fuels. Comparison was also conducted against their conjugate alkanes. Reaction path analysis was conducted to further investigate the oxidation process of the alkenes.

2. Experimental method

Figure 1a shows the schematic diagram of the experimental setup. A quartz tube with an inner diameter $d = 2$ mm, an outer diameter of 4 mm and a length of 200 mm was used as a reactor. Even though the inner diameter of the quartz tube is very small, wall chemical effect is confirmed to be negligible in the present experimental conditions [23,31,32]. A part of the quartz tube was heated from the bottom using a hydrogen/air premixed flat-flame burner to form a stationary wall temperature gradient of 300–1300 K in the flow direction as it is shown in the figure.

A gaseous pre-mixture with an equivalence ratio $\varphi = 1.0$ was introduced to the reactor with an inlet mean flow velocity $u = 2$ cm/s. Two kinds of mixture supplying methods were applied for gaseous and liquid fuels. For ethylene, propylene and 1-butene, which are gaseous at the standard condition, fuel/air pre-mixtures were used. The fuel and air were supplied separately using mass flow controllers (KOFLOC) to control equivalence ratio and inlet mean flow velocity independently. For 1-pentene, which is liquid at the standard condition, preparation of the gaseous mixture was done as follows. First, a stainless tank is evacuated using a vacuum pump and the liquid fuel was supplied using a syringe to obtain a specific pressure in the tank. And then nitrogen gas was added to the tank to form a gaseous fuel/N_2 mixture. The gaseous fuel/N_2 mixture was placed for a sufficient amount of time to assure the mixing. The gaseous fuel/N_2

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