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Study on combustion and ignition characteristics of natural gas components in a micro flow reactor with a controlled temperature profile

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ABSTRACT

Combustion and ignition characteristics of natural gas components such as methane, ethane, propane and *n*-butane were investigated experimentally and computationally using a micro flow reactor with a controlled temperature profile. Special attention was paid to weak flames which were observed in a low flow velocity region. The observed weak flame responses for the above fuels were successfully simulated by one-dimensional computations with a detailed kinetic model for natural gas. Since the position of the weak flame indicates the ignition characteristics as well as the reactivity of each fuel, the experimental and computational results were compared with research octane number (RON) which is a general index for ignition characteristics of ordinary fuels. At 1 atm, ethane showed the highest reactivity among these fuels, although RON of ethane (115) is between those of methane (120) and propane (112). Since the pressure conditions are different between the present experiment and the general RON test, weak flame responses to the pressure were investigated computationally for these fuels. The order of the fuel reactivity by the reactor agreed with that by RON test when the pressure was higher than 4 atm. Reaction path analysis was carried out to clarify the reasons of the highest reactivity of ethane at 1 atm among the employed fuels in this study. The analysis revealed that $C_2H_5 + O_2 \Leftrightarrow C_2H_4 + HO_2$ is a key reaction and promotes ethane oxidation at 1 atm. The effect of the pressure on the fuel oxidation process in the present reactor was also clarified by the analysis. In addition, weak flame responses to various mixing ratios of methane/*n*-butane blends were investigated experimentally and computationally. The results indicated a significant effect of *n*-butane addition in the blends on combustion and ignition characteristics of the blended fuels.

1. Introduction

Natural gas is a promising fuel to reduce emissions of CO_2 and NO_X. The use of natural gas is increasing in various fields as an energy source, e.g., gas turbines, boilers, furnaces, vehicle engines and so on. For improvement of combustion devices fueled by natural gas, knowledge on combustion and ignition characteristics of natural gas is significantly important. It is also known that the composition of natural gas varies with the region where it is produced, and the difference in composition significantly influences its combustion and ignition characteristics. Combustion and ignition characteristics of fuels related to natural gas have been extensively investigated in terms of flame structure [1,2], laminar burning velocity [3–6], ignition temperature [7], and ignition delay time [8–11]. There have been many studies on natural gas components, and many detailed reaction kinetic models for natural gas have been developed [12–15]. One of the recent detailed kinetic models consists of near 300 species and a thousand reactions and can be applied to C_1–C_5 alkane blends [15].

In this work, combustion and ignition characteristics of natural gas components were investigated using a micro flow reactor with a controlled temperature profile [16]. In the micro flow reactor system, a cylindrical quartz tube is employed as the reactor tube. The inner diameter of the tube is smaller than the ordinary quenching diameter. The tube is heated by an external heat source and a stationary temperature profile along the inner surface of the tube wall is formed in the flow direction. A premixed mixture is supplied to the reactor and the gas-phase temperature strongly depends on the given wall-temperature profile. In addition, the flow field in the reactor is laminar and at a constant pressure. These features make the reactor a simple system, which is a great advantage for the investigation of complicated combustion chemistry. In the previous study using the present reactor [16], three kinds of flames were observed: normal flame, FREI (Flames with Repetitive Extinction and Ignition) and weak flames, depending on the inlet mixture

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Nomenclature

A	cross-sectional area
c_p	specific heat at constant pressure
d	inner diameter of tube
h	specific enthalpy
M	mass flow rate
Nu	Nusselt number
P	pressure
T	temperature
U	mean flow velocity
V	diffusion velocity
W	molecular weight
x	spatial coordinate
Y	mass fraction
λ	thermal conductivity
ρ	density
ω	molar production rate

Subscripts

k	species index
w	wall

flow velocity. Normal flame is an ordinary stable flame and observed at a high velocity regime. FREI occurs at an intermediate velocity regime where the unstable middle-branch solution exists. At a low flow velocity regime, weak flames, which are stable and with quite weak chemiluminescence, were observed. This flame response to the flow velocity was also confirmed for methane [17], dimethyl ether (DME) [18], *n*-heptane [19], gasoline PRF (Primary Reference Fuel) [20] and diesel surrogates [21]. In recent works using the micro flow reactor, most of their attention was paid to weak flames because of the unique ability of this reactor to investigate combustion and ignition characteristics of the applied fuels. Tsuboi et al. investigated methane weak flame and found a lower inlet flow velocity limit of weak flames [17]. The wall temperature at the weak flame position corresponds to the minimum ignition temperature of the mixture in the given condition. Oshibe et al. investigated three-stage oxidation of DME in the reactor [18]. They observed two luminous zones which indicated separated hot flames (blue and hot flames). In addition to the two flames, they also confirmed low-temperature oxidation (cool flame) of DME by gas analysis. The three-stage oxidation of DME was reproduced by one-dimensional computation. Oxidation process of *n*-heptane in the reactor was investigated by Yamamoto et al. [19]. They observed three luminous zones of weak flame, which indicated cool, blue and hot flames. Note that a flame observed in the low velocity regime of the micro flow reactor is termed "weak flame" and each flame of separated weak flames is termed a cool, blue or hot flame depending on its oxidation process. In subsequent studies on gasoline PRF, Hori et al. investigated weak flame responses to research octane number (RON) with changing a mixing ratio of *n*-heptane and *iso*-octane [20]. With increasing RON, chemiluminescence from cool flame got weaker and the position of hot flame moved to the downstream side which is a higher temperature region. Diesel surrogate fuels which have different cetane numbers (CNs) were applied to the reactor by Suzuki et al. [21]. They confirmed that weak flame of a fuel which has lower cetane number stabilized in a higher temperature region. These results on weak flame response to RON and CN indicate that the positions of weak flames in the reactor represent the ignition characteristics, as well as the reactivity, of fuels and the existence of a strong relationship between general ignition indexes and the weak flame position. Since this reactor has unique characteristics, the results obtained by the micro flow reactor would be useful for further understanding of combustion and ignition characteristics of natural gas components and examining the validation of reaction kinetics.

In the present study, combustion and ignition characteristics of methane, ethane, propane and *n*-butane, which are the typical components of natural gas, were investigated using a micro flow reactor with a controlled temperature profile. The weak flames of these fuels were observed, and their positions were compared with the results of one-dimensional computation with a detailed kinetic model. The results of the fuel reactivity in the reactor were compared with their RONs. For further understanding the oxidation process of those fuels, reaction path analysis was conducted for the ethane and *n*-butane cases.

A weak flame response to various mixing ratios of methane/*n*-butane blends was also investigated using the present micro flow reactor experimentally and computationally. By changing the mixing ratio of the blends, the effect of relatively higher hydrocarbon elements in natural gas on combustion characteristics was studied.

2. Experimental method

Figure 1 shows a schematic of the experimental setup. A quartz tube with an inner diameter of 2 mm was employed as a reactor channel. This inner diameter was smaller than the ordinary quenching diameter of employed mixtures at the standard state. The quartz tube was heated by a hydrogen/air flat-flame burner and a stationary temperature profile from ambient temperature (about 300 K) to 1300 K was formed along the inner surface of the tube, as shown in Fig. 2. The maximum temperature was adjusted by controlling equivalence ratio and flow velocity of the hydrogen/air mixture. Hydrogen was chosen as a fuel of the external heating burner because of better observation of chemiluminescence from hydrocarbon flames in the reactor. Methane, ethane, propane, *n*-butane and methane/*n*-butane blends were used as fuels. A premixed fuel/air mixture at the stoichiometric condition was supplied into the reactor at an inlet mean flow velocity of 2 cm/s. Dried air compressed by an air compressor was used. The purities of fuels were as follows: methane (99.999%), ethane (99.99%), propane (99.99%) and *n*-butane (99%). Flow rate of a mixture (3.8 cc/min) was controlled using mass flow controllers and

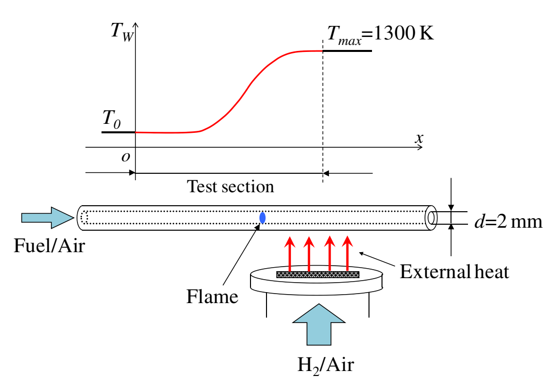

Fig. 1. Schematic of experimental setup.