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Laminar burning velocity and Markstein length of ammonia/air premixed flames at various pressures

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HIGHLIGHTS

- Laminar burning velocities of ammonia/air flames at high pressures are evaluated.
- Maximum value of laminar burning velocity of ammonia/air flame is about 7 cm/s.
- Laminar burning velocity decreases with the increase in the pressure.
- Markstein length increases with the increase in equivalence ratio.
- Markstein lengths at high pressure are lower than those at 0.1 MPa.

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ABSTRACT

Ammonia is expected to be useful not only as a hydrogen-energy carrier but also as a carbon-free fuel. In order to design an ammonia fueled combustor, fundamental flame characteristics of ammonia must be understood. However, knowledge of the characteristics of ammonia/air flames, especially at the high pressures, has been insufficient. In this study, the unstretched laminar burning velocity and the Markstein length of ammonia/air premixed flames at various pressures up to 0.5 MPa were experimentally clarified for the first time. Spherically propagating premixed flames, which propagate in a constant volume combustion chamber, were observed using high-speed schlieren photography. Results indicate that the maximum value of unstretched laminar burning velocities is less than 7 cm/s within the examined conditions and is lower than those of hydrocarbon flames. The unstretched laminar burning velocity decreases with the increase in the initial mixture pressure, tendency being the same as that of hydrocarbon flames. The burned gas Markstein length increases with the increase in the equivalence ratio, the tendency being the same as that of hydrogen/air flames and methane/air flames. The burned gas Markstein lengths at 0.1 MPa are higher than those at 0.3 MPa and 0.5 MPa. However, the values of burned gas Markstein length at 0.3 MPa and 0.5 MPa are almost the same. In addition, numerical simulations using CHEMKIN-PRO with five detailed reaction mechanisms which are presently applicable for the ammonia/air combustion were also conducted. However, qualitative predictions of unstretched laminar burning velocity using those reaction mechanisms are inaccurate. Thus, further improvements of reaction mechanisms are essential for application of ammonia/air premixed flames.

1. Introduction

Ammonia holds promise as a hydrogen energy carrier because of its high hydrogen weight of 17.7% [1]. At present, ammonia is widely used as a fertilizer. Application of ammonia as a hydrogen energy carrier would be useful because the infrastructure for ammonia distribution has already been established and the ammonia production process, i.e., the Haber–Bosch process, is well-known. Recently, a new process for producing ammonia by utilizing renewable energy sources, such as solar energy, is being studied [2]. In addition, ammonia also shows promise as a carbon-free fuel. However, in general, because of its lower combustion intensities, which means lower calorific value, slower laminar burning velocity and narrower flammability range, ammonia has not been considered as a fuel. Hence, fundamental flame characteristics of ammonia flame have been insufficiently studied.

In order to improve the lower combustion intensities of ammonia, hydrogen-added ammonia flames have been studied [3–7]. Lee et al. [3,4] experimentally investigated the laminar burning

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velocity of hydrogen-added ammonia/air premixed flames. They showed the laminar burning velocity and Markstein numbers at the atmospheric pressure and clarified that the laminar burning velocity increases and the Markstein number decreases with an increase in hydrogen concentration. Kumar and Meyer [5] showed the laminar burning velocity of hydrogen/ammonia/air flames by considering the heat loss from the flames. The application of ammonia in an actual combustor is also being studied. Frigo et al. [6,7] applied hydrogen-doped ammonia as a fuel for the spark ignition engines. The power and thermal efficiencies of a hydrogen-doped ammonia fielded engine were compared with those of a gasoline engine. In addition, consideration is being given to on-board cracking of ammonia in order to obtain hydrogen as a flame promoter in SI engines.

Recently, flame characteristics of ammonia/air premixed flames have been studied. Hayakawa et al. [8] experimentally and numerically investigated premixed ammonia/air laminar flames at atmospheric pressure as well as at elevated pressures. Experimental and numerical observations showed that the NO formation from ammonia/air flames was reduced in rich mixtures because of the reduction NO by ammonia. In addition, it was also clarified that NO formation decreases with an increase in pressure due to the enhancement of consumption of H and OH radicals, which play an important role in NO formation.

Detailed reaction mechanisms of ammonia flames have been developed by Miller et al. [9] and Lindstedt et al. [10], and reaction mechanisms of C reactions, including N chemistry, are also available for ammonia combustion as proposed by Konnov [11] and Tian et al. [12]. However, since these mechanisms were not elucidated based on ammonia/air flames, it is unclear whether they are applicable for ammonia/air flames owing to insufficiency of experimental results for ammonia/air combustion.

Laminar burning velocity is one of the most important parameters for premixed flames and for confirmation of reaction mechanisms. The laminar burning velocities of ammonia/air premixed flames have been investigated by Zakaznov et al. [13], Takizawa et al. [14] and Pfahl et al. [15]. According to their results, the laminar burning velocity of ammonia/air premixed flames is lower than that of hydrocarbon flames, such as methane/air flames and propane/air flames. In studies of Takizawa et al. [14] and Pfahl et al. [15], spherically propagating flames in a constant volume combustion chamber were investigated for the evaluation of laminar burning velocity. Although flame stretch, which causes a change in flame speed owing to thermo-diffusive effects [16], was known to occur for spherically propagating laminar flames, the effects of flame stretch on flame speed were not considered in these studies. In addition, all experiments were conducted at atmospheric pressure.

Spherically propagating premixed flames in a constant volume combustion chamber have been widely investigated in order to clarify the fundamental characteristics of premixed flames [17–20]. Kitagawa and Hayakawa et al. [17,18] investigated spherically propagating laminar and turbulent flames in a large constant volume combustion chamber for hydrogen and iso-octane up to initial mixture pressure of 0.5 MPa. Tse et al. [19] investigated flame propagation up to 60 atm using two concentric cylindrical vessels. Kelley et al. [20] proposed a non-linear relationship between flame speed and flame stretch rate.

The Markstein length and the Markstein number are also important parameters for laminar flames because these values express the role of thermo-diffusive effects. Although these values are important for laminar flame characteristics, such as flame instability [21], turbulent flame characteristics are also affected by the Markstein length or the Markstein number. Hayakawa et al. [18] investigated the turbulent burning velocity at fixed turbulence Karlovitz numbers and showed that the ratio of turbulent burning velocity to unstretched laminar burning velocity increases with a decrease in Markstein number. Kobayashi et al. [22] proposed that the Markstein length affects the flame surface density of the turbulent flame front via a change in the passivity of the flame front. Bradley et al. [23] expressed the correlation between turbulent burning velocity and effective turbulence intensity. Therefore, evaluation of the Markstein length or the Markstein number is also important for an understanding of turbulent combustion.

The objectives of the present study were to evaluate the laminar burning velocity and the Markstein length of ammonia/air premixed flames at various pressures up to 0.5 MPa. The equivalence ratios were varied from 0.7 to 1.3. In addition, numerical simulations using various detailed reaction mechanisms were also carried out. The results of numerical simulations are compared with the experimental results in the next sections.

2. Experimental setup and numerical method

Fig. 1 shows the experimental setup used in this study. Experiments were carried out using a constant volume cylindrical combustion chamber. The inner diameter and length of the chamber were 270 mm and 410 mm, respectively. The volume of the chamber was approximately 23 L, equivalent to that of a sphere with a diameter of 355 mm. The mixture was ignited by spark electrodes near the center of the chamber. The diameter of the spark electrodes was 1.5 mm and the spark gap was set to 2 mm.

A capacitor discharge ignition (CDI) circuit was adopted for spark ignition of the mixture. The electrostatic energy, which was charged in the capacitor in the CDI circuit, was set to 2.8 J. Although the energy of 2.8 J was different from the ignition energy and extremely larger than the minimum ignition energy of usual fuels, such as hydrocarbons [24], it was required to ensure the ignition of the ammonia/air mixture at atmospheric pressure. As described in Section 4.2, the ignition-affected regimes during flame propagation were not considered in the analysis of the determination of unstretched flame speed and Markstein length. Optical windows made of quartz glass were located opposite each other and flame propagation could be observed via these two windows.

Ammonia was used as the fuel and air was used as the oxidizer. Experimental conditions are summarized in Table 1. Initial mixture pressure, P_i, and the equivalence ratio, ϕ, were varied from 0.1 to 0.5 and 0.7 to 1.3, respectively. All experiments were conducted at a temperature of 298 K. The mixture was prepared according to the partial pressure of ammonia and air. The pressure inside of the chamber was measured by a pressure sensor (P1 in Fig. 1, GE Sensing UNIC5000). Experiments were carried out at least five times for each examined condition. The unstretched laminar burning velocity and burned gas Markstein length, which will be described in Section 4.2, were determined as the averaged value of the results obtained from each experiment. Experimental fluctuation was defined as the difference between maximum and minimum values of the experimental results. Schlieren photography with a high-speed camera and a continuous light source was adopted for the flame observation. A metal halide lamp (Photron, HVC-SL) and a high-speed camera (Photron, FASTCAM SA5) were used for the schlieren system. In order to form a spotlight source, a pinhole was mounted in front of the metal halide lamp. A micro lens (Nikon, Ai AF Micro-Nikkor 200mm f/4D IF-ED) was mounted on the high-speed camera. The resolution of the schlieren photography was 768 × 768 and the frame rate was 1000 fps. The resolution of the schrieren images was approximately 0.1 mm/pixel. Flame propagation could be obtained up to 60 mm in diameter using the schlieren technique via two optical windows. In addition, direct color high-speed photographs were taken by a high-speed