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Combustion and ignition characteristics of ammonia/air mixtures in a micro flow reactor with a controlled temperature profile

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Abstract

Combustion and ignition characteristics of a stoichiometric ammonia/air mixture were investigated by a micro flow reactor with a controlled temperature profile from ambient temperature to 1300 K at atmospheric pressure. Three kinds of flame dynamics depending on inlet mean flow velocities, namely, normal flames in the high velocity regime, flames with repetitive extinction and ignition (FREI) in the intermediate velocity regime, and weak flames in the low velocity regime, which have also been observed for hydrocarbons in previous studies, were observed for ammonia by the direct observation with a digital still camera in this study. The existence of ammonia weak flames at 1270 K was confirmed. Special attention was paid to weak flames to examine ignition characteristics of an ammonia/air mixture at low temperature because ignition experiments of ammonia have been conducted only at very high temperatures (around 2000 K). Species measurement for a stoichiometric ammonia/air weak flame at 10 cm/s was made using and a mass spectrometer and a T-shaped reactor fused with a micro-probe. The present species measurement elucidated the structure of the ammonia/air weak flame and complete combustion was confirmed at 1290 K. Five reaction mechanisms were used for computations of weak flames and the four of them did not predict complete combustion within the given computational domain. One reaction mechanism predicted complete combustion but the weak flame position in computation was located in a temperature region lower than that in the experiment. The order of reactivity evaluations among the five mechanisms using weak flames in the micro flow reactor agreed with that using ignition delay times at low temperatures which are generally difficult to be taken by ignition experiments. The capability of weak flame methodology to validate ignition properties of reaction mechanisms for low reactivity fuels like ammonia was successfully demonstrated in this study.

Keywords: Ammonia; Nitrogen oxides (NO*x*); Microcombustion; Energy carrier; Chemical kinetics

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1. Introduction

Ammonia (NH_3) is attracting attention as a potential hydrogen carrier because of its high hydrogen density (17.7 wt%) and favorable characteristics of liquefaction for storage and transportation (similar to propane) [1–3]. NH_3 direct combustion has potential for utilization of an energy carrier because NH_3 has a great advantage as a carbon-free fuel. However, the reactivity of NH_3 is much lower than those of hydrocarbons, and fundamental knowledge of combustion and ignition characteristics of NH_3 are quite limited compared with that of hydrocarbons. To apply existing combustion technologies and combustors for NH_3, accurate and reliable combustion models of NH_3 need to be developed through extensive validations using fundamental combustion and ignition data of NH_3.

Since NH_3 is a good reductant in the denitration process, NH_3 has been widely used in combustion facilities to reduce NOx emission. Fundamental studies on reduction reactions of NOx by NH_3 have been extensively [4–7] performed. In addition, NH_3 is a representative nitrogen compound included in practical fuels, especially in biomass and coal, and is an important source of fuel NOx. Therefore, NOx formation reactions from NH_3 have also been studied [8–11]. The NOx reaction system is a sub-mechanism of the NH_3 oxidation and NOx formation mechanisms have been utilized in the development of NH_3 reaction mechanisms [12–16]. The developed NH_3 reaction mechanisms have been validated using experimental results on laminar flame speeds [17–22] and ignition delay times [23–28]. However, fundamental combustion and ignition data of NH_3 are still limited. Especially for ignition data, ignition delay times have been measured for NH_3 mixed with CH_4 or H_2, or $NH_3/O_2/Ar$ mixtures only in very high temperature conditions using shock tubes (around 2000 K). Recent studies have reported that ignition characteristics at low-to-intermediate temperatures play an important role in the base of premixed turbulent jet flames [29], and longer residence time by introducing swirl and recirculation near the base of premixed turbulent jet flames has successfully attained stable combustion with NH_3/air mixtures [30]. Therefore, ignition characteristics at lower temperatures for NH_3/air mixtures need to be examined for future development of NH_3 combustors. However, experiments of ignition delay times for NH_3/air mixtures at lower temperatures (around 1200 K) are quite difficult due to the very long ignition delay times of such mixtures.

This study employed a different methodology, a micro flow reactor with a controlled temperature profile [31–42], to investigate ignition characteristics of NH_3/air mixtures at low temperatures. Three kinds of flame dynamics have been observed in the micro flow reactor depending on the inlet mean flow velocity: steady normal flames in the high velocity regime, unsteady flames with repetitive ignition and extinction (FREI) in the intermediate velocity regime, and steady weak flames in the low velocity regime [31]. Particular attention has been paid to weak flames to investigate ignition characteristics of given fuels because theoretical analysis has shown that the weak flame branch is on the ignition branch of the Fendell curve [32]. The transient, two-stage ignition, which is commonly observed for hydrocarbons having the low-temperature oxidation, has been observed as steady, three-stage weak flames in the micro flow reactor (low-temperature oxidation and separated high-temperature oxidation) [33,34]. Fuel reactivity has been successfully evaluated for gasoline primary reference fuels (PRFs) [35], diesel PRFs [36], natural gas components [37] and C2–C5 alkenes [38] by the weak flame position as an index. Weak flames of higher/lower reactivity fuels have been found to be located in the lower/higher temperature region. The gas-phase temperature profile is nearly equal to the given wall-temperature profile even in the reaction zone in the weak flame regime [39] and the steady, 1-D numerical model of the micro flow reactor has enabled the validation of the employed reaction mechanisms under the well-defined temperature profile by comparison with experimental results [40–42].

This study aimed to apply the micro flow reactor for NH_3/air mixtures to investigate their ignition properties at low temperatures (around 1200 K) because long residence time under the controlled temperature profile in the micro flow reactor is suitable for experiments of low reactivity fuels like NH_3. Flame dynamics in the micro flow reactor for stoichiometric NH_3/air mixtures were investigated and the existence of weak flames for NH_3 was confirmed. Species measurement for the NH_3 weak flame was then made by a mass spectrometer and numerical simulations with various NH_3 reaction mechanisms were made to compare them with experimental results.

2. Experimental setup

Figure 1 shows a schematic of the micro flow reactor. A quartz tube with an inner diameter of 2 mm was used as a reactor channel and was heated by a hydrogen/air flat-flame burner to give a stationary temperature ramp from ambient temperature to 1300 K along the inner surface of the reactor in the flow direction. Hereafter, "wall temperature" means the temperature on the inner surface of the reactor. The wall-temperature profile in the flow direction was measured using a K-type thermocouple (junction diameter of 0.25 mm)

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