Properties of Nonpremixed Ammonia-substituted Hydrogen-air Flames

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ABSTRACT

The extinction and nitrogen oxide (NO_x) emissions of nonpremixed ammonia (NH_3)-hydrogen (H_2)-air flames were experimentally studied to evaluate the potential of partial NH_3 substitution to improve the safety of H_2 use and provide a database of nonpremixed NH_3-substituted H_2-air flames. Considering coflow nonpremixed NH_3-H_2-air flames at normal temperature and pressure for a wide range of fuel and coflow air injection velocities and the extent of NH_3 substitution, the effects of NH_3 substitution and fuel and air injection velocities on the combustion stability (extinction) limits and NO_x emissions of H_2-air flames were investigated. Results show reduction of stability limits with NH_3 substitution, supporting the potential of NH_3 as an effective, green (i.e., carbon-free) additive in H_2-air flames. The NO_x emission index (EI_{NOx}) increases with enhanced NH_3 substitution in general. For a given air injection velocity, EI_{NOx} decreases and then increases with increasing fuel injection velocities.

Keywords: ammonia, hydrogen, nonpremixed flames, stability limits, carbon-free

1 INTRODUCTION

In spite of increasing interest in the use of pure hydrogen (H_2) in combustion devices due to its unique features such as no carbon dioxide (CO_2) emissions and wide flammability limits [1,2], the inherently high diffusivity and reactivity of H_2 are responsible for the potential of explosion hazards in storage. Also, non-luminous H_2 flames are difficult to be visually detected and optically monitored. Some methods to suppress hazards due to H_2 usage such as the use of hydrocarbon-substituted H_2 in combustion devices have been developed [3]. Recently, the use of carbon-free ammonia (NH_3)-H_2 mixtures in combustion devices as an alternative approach towards enabling H_2 utilization and diminishing the associated safety problems have been proposed [4–6]. Similar to reaction of pure H_2-air mixtures, reaction of NH_3-H_2-air mixtures does not produce CO_2, while NH_3 is easily storable, which is different from H_2 [7]. Although some technological challenges and public concerns are imposed by the use of NH_3 [8], potentially beneficial characteristics of the combustion of NH_3-H_2-air mixtures have been discussed in the previous study [4]: e.g., diminished the propensity of the mixtures to detonate due to the presence of NH_3 in H_2-air flames and enhanced the safety of H_2 use due to visual detection of luminous NH_3-H_2 flames.

In the previous studies in this laboratory [4–6], the potential of NH_3 as a carbon-free, green additive for improving the safety of H_2 use in both premixed and nonpremixed H_2-air flames was observed. The experimental and computational study on the propagation and nitrous oxide (N_2O) and nitrogen oxide (NO_x) emissions of spark-ignited spherical laminar premixed NH_3-H_2-air flames shows substantial reduction of laminar burning velocities with NH_3 substitution in H_2-air flames [4]. The potential of NH_3 as a suppressant of both hydrodynamic and preferential-diffusional cellular instabilities in premixed H_2-air flames was also found. Although NH_3 substitution enhances the N_2O and NO_x formation in general, the increased amount of N_2O and NO_x emissions with NH_3 substitution for fuel-rich conditions is much lower than that under fuel-lean conditions. The computational study of the effects of NH_3 substitution on the extinction limits and structure of counterflow nonpremixed H_2-air flames shows reduction of the blowoff limits, the concentration of light radicals and the maximum flame temperature with NH_3 substitution in H_2-air flames [5]. It was found that chemical effects than thermal effects of NH_3 substitution on flame structure are dominant and radiation effects on the extinction limits and flame structure are not remarkable particularly for high-stretched flames. The most recent study of burner-stabilized premixed NH_3-H_2-air flames also supports the potential of NH_3 as a carbon-free additive for improving the safety of H_2 use, showing a reduction of stability limits with NH_3 substitution and coflow [6]. It was observed that the thermal $deNO_x$ process in the post-flame region is involved in reducing NO_x emissions for fuel-rich flames.

The aim of the present investigation is to extend previous work [4–6] on the potential of improving H_2 safety by adding NH_3 to H_2-air flames and provide a useful database for modeling nonpremixed NH_3-H_2-air flames, with the following specific objectives. The first is to measure the combustion stability (extinction) limits of nonpremixed NH_3-H_2-air flames in order to quantify the extent of stability limit reduction due to NH_3 addition. The second is to measure the NO_x emissions of nonpremixed NH_3-H_2-air flames in a closed chamber in order to observe the effects of NH_3 substitution and fuel and coflow air injection velocities on NO_x emissions. Thus, this study has been conducted for a wide range of fuel and air injection velocities and the extent of NH_3 substitution.

2 EXPERIMENTAL METHODS

The nonpremixed flame in a coflow using a tube type burner was adopted for the present investigation because it is the most common flame configuration for studying the combustion stability (extinction) limits and NO_x emissions of nonpremixed flames. The experimental apparatus we employed is shown schematically in Fig. 1. It consists of a fuel tube surrounded with a concentric tube to provide coflow air, a cylindrical lab-scale combustion chamber that is connected to the fuel and coflow air delivery tubes, a gas analyzer for measuring NO_x emissions, thermocouples (R-type) for measuring temperature distribution in the flame and a digital camera (Kodak Z812IS) for recording flame images.

A NH_3-H_2 mixture is injected from a stainless steel (SUS316L) tube with inner and outer diameters of 3.00 and 5.26 mm, respectively, and with the passage length/ diameter ratio of 150 to help insure fully-developed pipe flow at the tube exit. Coflow air is injected from a concentric stainless steel (SUS316L) tube with an inner diameter of 15.0 mm, while ambient air at low velocities is provided through the honeycomb plate that is the bottom (upstream) of the cylindrical combustion chamber (plexi-glass) with the inner diameter, height and thickness of 210, 960 and 8 mm, respectively. The coflow nonpremixed jet is spark-ignited so that a nonpremixed flame is formed and stabilized on the tube rim. Flow meters (CSM: 0–450 lpm) with accuracy ±3.0% of full scale deliver the fuel mixture and air to the main and concentric tubes, respectively. Using the flow meters, fuel-mixture composition (the mole fraction of NH_3 in the fuel gas $x_a = X_{NH3}/(X_{NH3} + X_{H2})$ where X_{NH3} and X_{H2} are the mole fractions of NH_3 and H_2, respectively) and tube exit velocity V_{fuel} and coflow air tube exit velocity V_{coflow} are independently controlled. To avoid bending or melting due to hot product gases, the top cover of the combustion chamber is made of aluminum, and the exhaust port that is located on the center of the chamber top plate is connected directly to a ventilation tube.

The concentration of NO_x was measured in the ventilation tube using the gas analyzer (Testo 350-XL) with an accuracy of 0.1–1.0 ppm, the probe of which is located on the center of the ventilation path. The combustion stability limits were measured as follows. As aforementioned, NH_3-substituted H_2-air nonpremixed flames were obtained by establishing a cold coflow jet of fuel mixture and air and then igniting the coflow jet with a spark. With and without coflow air the ambient air was always injected at 1.2 m/sec. For all experiments, the concentrations of NH_3 and H_2 (i.e., x_a) were set to fixed values in each mixture and then V_{coflow} was varied to find the combustion stability limits for a given value of V_{fuel}. Given V_{fuel} and x_a, upper combustion stability limits were observed at high V_{coflow} due to the fuel-lean mixture concentration in the flame base (blowout). In the present study, however, we have not measured lower combustion stability limits at low V_{coflow} because of the limited capability (i.e., flow meter resolution) of the present apparatus, just focusing on the blowout limits. The bowout limits were obtained by establishing a stable flame at a relatively lower V_{coflow} and then gradually increasing V_{coflow}. Since flames were lifted off at high V_{fuel}, the liftoff limits were also measured. Final results, including the combustion stability limits and NO_x emissions of nonpremixed NH_3-substituted H_2-air flames, were obtained by averaging measurements of 4 to 6 tests at each condition. Experimental uncertainties (95% confidence) for V_{fuel}, V_{coflow} and x_a were less than 5%. At temperature $T = 298\pm3$ K and atmospheric pressure (NTP) experiments were carried out for x_a = 0.2–0.4, V_{fuel} = 23.6–235.8 m/s (Reynolds number Re = 619–6,193) and V_{coflow} = 0–13.7 m/s.
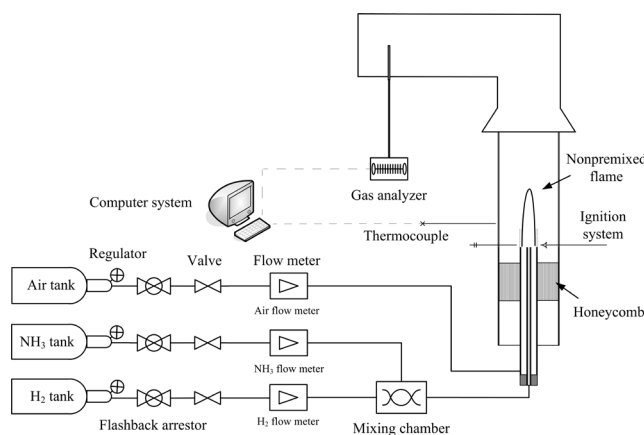

Figure 1: Schematic of experimental apparatus.

3 RESULTS AND DISCUSSION

3.1 Combustion Stability Limits

Effects of NH_3 substitution on the combustion stability limits were examined to provide a database for modeling nonpremixed NH_3-substituted H_2-air flames and to quantify the extent of stability limit reduction due to NH_3 addition under various fuel mixture injection conditions and thereby confirm the potential of partial NH_3 substitution to improve the safety of H_2 use without losing the unique feature of no CO_2 emissions for pure H_2 use.

Figure 2 shows the measured combustion stability limits, including the blowout and liftoff limits, on a V_{coflow}-V_{fuel} diagram for nonpremixed NH_3-H_2-air flames with a fixed x_a = 0.4 at NTP. Upper combustion stability (blowout) limits are observed at high V_{coflow} since the mixture in the flame base becomes fuel-lean and thus, the flame becomes weak and is finally extinguished with relativly high local strain rates. Up to V_{fuel} = 88.4 m/sec no liftoff flame is observed, while beyond V_{fuel} = 88.4 m/sec flames are lifted off at low V_{coflow} since the position where the local flow velocity matches the local burning velocity shifts downstream with enhanced V_{fuel}, i.e., the local strain rates near the burner exit are too high to match the local burning velocities [9]. A

typical extinction behavior of nonpremixed flames is observed: V_{coflow} at the extinction increases (decreases) with increasing V_{fuel} for laminar (turbulent) NH_3-H_2-air flames. Thus, with increasing V_{fuel} the range of combustion stability limits becomes extended until V_{fuel} = 47.2 m/sec and then reduced.

Figure 3 shows the combustion stability limits for nonpremixed NH_3-H_2-air flames with a fixed x_a = 0.2 at NTP. They can be compared with the results for x_a = 0.4 in Fig. 2, showing the effects of the extent of NH_3 substitution on the combustion stability limits. Similar to the results for x_a = 0.4, the blowout and liftoff limits are observed for high V_{fuel}, while only the blowout limits are observed for low-to-moderate V_{fuel}. Compared with the combustion stability limits for x_a = 0.4, they have been substantially extended, showing that V_{coflow} at the blowout limits increases. This tendency is observed because the burning intensities of NH_3-H_2-air flames, i.e., the local burning velocities of NH_3-H_2-air mixtures, have been enhanced with lower NH_3 substitution. Due to the enhanced burning intensity the V_{fuel} limit has been also extended: V_{fuel} = 23.6–235.8 m/sec compared with 23.6–94.3 m/sec for the flames of x_a = 0.4 (Fig. 2).
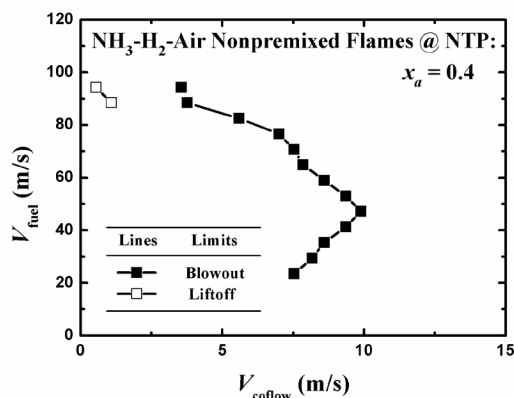

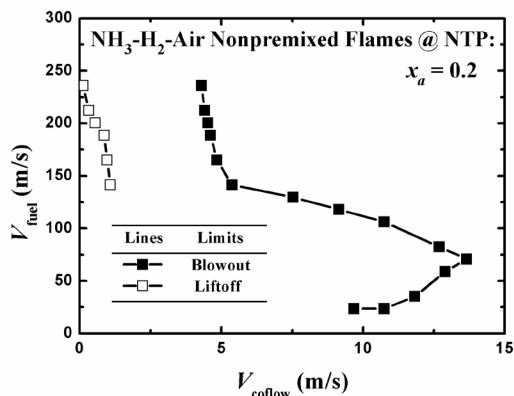Figure 2: Stability limits of nonpremixed NH_3-substituted H_2-air flames of x_a = 0.4 at NTP.

Figure 3: Stability limits of nonpremixed NH_3-substituted H_2-air flames of x_a = 0.2 at NTP.

The results in Figs. 2 and 3, showing reduction of the combustion stability limits with NH_3 substitution in H_2-air flames, support the potential of NH_3 as a carbon-free, green additive for improving the safety of H_2 use in nonpremixed H_2-air flames in addition to the previous results from premixed and counterflow nonpremixed NH_3-substituted H_2-air flames at NTP [4–6].

3.2 NOx Formation

Effects of NH_3 substitution, V_{fuel} and V_{coflow} on the NO_x emissions of nonpremixed NH_3-H_2-air flames were examined to evaluate the performance of combustion devices using NH_3-H_2 mixtures and the results are given in Figs. 4 and 5. The NO_x emission index EI_{NOx} that is defined as the fraction of the mass (g) of the produced NO_x per the mass (kg) of the provided NH_3 and H_2 is used as a performance parameter. In addition to the NO_x emission data, the NO data are separately provided.

Figure 4 shows EI_{NOx} for nonpremixed NH_3-H_2-air flames with x_a = 0.4 and V_{coflow} = 0 and 2.2 m/s at NTP. It is observed that for V_{coflow} = 0 (only with the ambient air) EI_{NOx} decreases up to V_{fuel} = 70.7 m/s and then increases with increasing V_{fuel}, though the absolute value of NO_x emissions monotonically increases with increasing V_{fuel} (not shown here). The tendency of EI_{NOx} decrease with increasing V_{fuel} for low V_{fuel} is observed since the effects of recirculation of burned gas in the combustion chamber that encloses the coflow burner dilute unburned mixtures for low V_{fuel} where flame is relatively small [10]. Meanwhile, the tendency of EI_{NOx} increase with increasing V_{fuel} for high V_{fuel} is observed since the reduced flame length (residence time) due to the liftoff behavior and thereby the reduced radiant heat losses enhance NO_x emissions for high V_{fuel} where flame is relatively large and thus the effects of recirculation of burned gas are not enhanced any more. When coflow air is injected (V_{coflow} = 2.2 m/s), EI_{NOx} has been somewhat reduced due to the dilution effects of added coflow air.

Figure 5 shows EI_{NOx} for nonpremixed NH_3-H_2-air flames with x_a = 0.2 and V_{coflow} = 0 and 2.2 m/s at NTP. They can be compared with the results for x_a = 0.4 in Fig. 4, showing the effects of the extent of NH_3 substitution on the NO_x emissions. Similar to the results for x_a = 0.4, EI_{NOx} decreases up to a certain value of V_{fuel} (= 94.3 m/s) and then increases with increasing V_{fuel} for no coflow condition (only with the ambient air), while EI_{NOx} is reduced with coflow air. Compared with EI_{NOx} for x_a = 0.4, it has been somewhat reduced, showing that EI_{NOx} increases with enhanced NH_3 substitution, though it is not remarkable. Meanwhile, the flame temperature decreased with increasing NH_3 substitution (though data is not provided). Thus, this result indicates that the thermal NO_x mechanism does not play the main role in enhancing NO_x emissions with increasing NH_3 substitution for the nonpremixed NH_3-H_2-air flames.

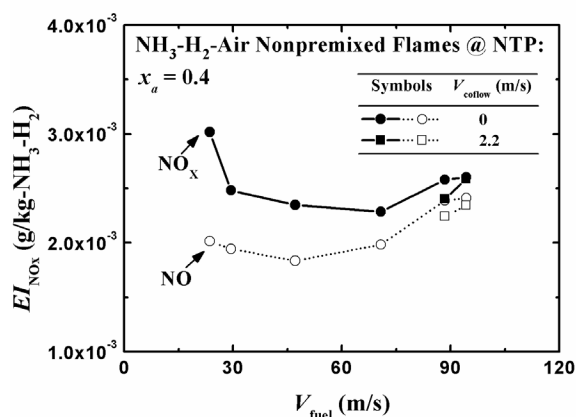Figure 4: Effects of V_{fuel} and V_{coflow} on NO$_x$ emission index of nonpremixed NH$_3$-substituted H$_2$-air flames of $x_a = 0.4$ at NTP.

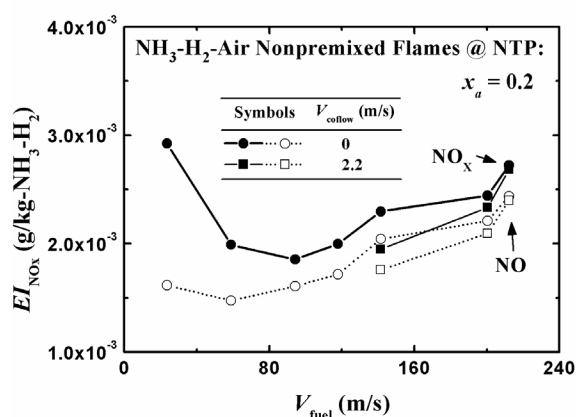Figure 5: Effects of V_{fuel} and V_{coflow} on NO$_x$ emission index of nonpremixed NH$_3$-substituted H$_2$-air flames of $x_a = 0.2$ at NTP.

4   CONCLUSIONS

The effects of NH$_3$ substitution on the combustion stability (extinction) limits and NO$_x$ emissions of nonpremixed H$_2$-air flames at normal temperature and pressure were experimentally studied to evaluate the potential of partial NH$_3$ substitution to improve the safety of H$_2$ use and provide a database of nonpremixed NH$_3$-substituted H$_2$-air flames. The major observations and conclusions of the study are as follows. A typical extinction behavior of nonpremixed flames is observed: the blowout limits at high coflow air tube exit velocity V_{coflow} due to the fuel-lean mixture concentration in the flame base and the liftoff limits at low V_{coflow} due to the high local strain rates near the burner exit compared with the local burning velocities. The coflow air tube exit velocity at the extinction increases (decreases) with increasing fuel tube exit velocity V_{fuel} for laminar (turbulent) NH$_3$-H$_2$-air flames.

Ammonia substitution substantially reduces the blowout limits. The NO$_x$ emission index EI_{NOx} increases with enhanced NH$_3$ substitution, though it is not remarkable. For given V_{coflow} EI_{NOx} decreases and then increases with increasing fuel tube exit velocity, though the absolute value of NO$_x$ emissions monotonically increases. The effects of recirculation of burned gas in the combustion chamber that encloses the coflow burner reduce EI_{NOx} for low V_{fuel}, while the reduced flame length due to the liftoff behavior and thereby the reduced radiant heat losses enhance EI_{NOx} for high V_{fuel}. The present observations support the potential of NH$_3$ as a carbon-free, green additive for improving the safety of H$_2$ use with reasonable levels of NO$_x$ emissions.

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