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Laminar flame speeds of ammonia mixtures at high pressure and temperature conditions: New experimental results and performance of different kinetic models

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ABSTRACT

There is a need for alternative sources of energy to meet with the ever-increasing demand leading to a rapid depletion of fossil fuels as well as to reduce carbon emissions which is the primary cause of global warming. Ammonia stands as a potential fuel as it is carbon-free, easily available, and can be stored and transported safely. Studies show that it is a viable fuel that can be used in spark-ignition engines and gas turbines. The goal of this study is to provide experimental flame speeds for extreme conditions encountered in engines and also to evaluate the most recent kinetic mechanisms.

1. Introduction

In modern times, the energy demand has increased leading to an extensive use of hydrocarbon-based fuels. This has not only led to the overconsumption of the resources, but also has augmented the amount of greenhouse gases and pollutants in the atmosphere that are responsible for climate change. Ammonia is carbon-free which makes it a prospective contender among other green fuels. Moreover, it can be safely stored and transported and can be manufactured in large quantities by the Haber-Bosch process. There are some limitations to using pure ammonia as a fuel due to its flammability limit ($\varphi = 0.63 - 1.4$)[1] and low heating value (18.8 *MJ/kg*) [1]. However, it has an anti-knock characteristic due to its high-octane number making it favorable to use in spark-ignition engines and gas turbines [2]. Ammonia-air flames are characterized by low flame speeds; therefore, ammonia is often combined with hydrogen or methane to improve its combustion [2]. Literature data is available for these but not at elevated conditions which are encountered in engines and gas turbines. This study aims to perform experiments at high temperatures and high pressures in constant volume conditions for different equivalence ratios to provide laminar flame speeds and further, to evaluate the capabilities of the chemical kinetic mechanisms available in the literature.

2. Method

2.1 Experimental set up

One of the most common and accurate methods of measuring the flame speeds consists in observing a spherical flame propagating outward in a constant volume chamber. This method enables to make measurements in extreme conditions. The experiments are performed in the OPTIPRIME facility [3] of ICARE-CNRS, Orléans, France. It is a novel constant volume combustion chamber which allows a full radial visibility as it uses a 360° fused silica ring. The flame front can be tracked from its ignition point till it reaches the wall. The idea behind the vast visibility range is to be certain that the flame front remains spherical throughout its propagation without being influenced by gravity effects or instabilities. The apparatus is fitted with K-type thermocouple for initial temperature measurement and pressure transducers (AVL GU21D) to measure pressure simultaneously with the flame radius evolution.

2.2 Experimental conditions and measurement of flame speeds

Mixtures of ammonia and oxygen at three different equivalence ratios: 0.8, 1.1 and 1.3 at an initial temperature of 300 K have been studied. The oxy-ammonia flames are quite unstable as both hydrodynamic and thermo-diffusive instabilities are favored at high temperatures and pressures. In order to stabilize these flames, it is necessary to add a gas with high thermal diffusivity to the mixture. A combination of argon and helium was used to get stable flames. The ratio of the diluents was chosen such that the mixture can be ignited easily and the flame can sustain. Helium possesses a high thermal diffusivity. A mixture containing a large percentage of helium is used only for the high-pressure initial conditions as larger instabilities are incurred in these conditions. Oxygen constitutes 30% of the oxidizer mixture at all the tested conditions. The remaining 70% consists of the diluents. The initial pressure for the rich mixtures ranged between 1 and 4 bar whereas, for the lean mixtures, the initial pressure was set at 1 and 2 bar. Using the isochoric method allows obtaining a wide band of flame speed data at elevated conditions of both pressure and temperature. It is assumed that the temperature of fresh gases evolves isentropically while the flame front propagates. Since the evolution is isentropic, the effect of radiation heat losses on the flame speed is negligible. In the given experimental set-up, the data has been limited to $2 < \frac{P}{P_0} < 5$ [3]. This range has been selected so that stretch effects occurring at the initial phase and heat losses endured when the flame front gets closer to the wall do not affect the propagation of the front and hence, the flame speed. Table 1 represents the initial pressure and the equivalence ratio along with the range of pressure and temperature for which the flame speed data can be measured. The maximum data available in the literature is for a pressure at 5 bar and for temperature at 473 K. In the current study, 20 bar and 485 K can be reached.

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Table 1 Initial pressure and equivalence ratio with the range of pressure and temperature for which the flame speed has been calculated. The initial temperature, T_0, was maintained at 300 K.

φ	P_0(bar)	P range (bar)	T range (K)
0.8	1	2-5	368-485
0.8	2	4-10	366-484
1.1	1	2-5	369-482
1.1	2	4-10	371-485
1.1	3	6-15	369-482
1.1	4	8-20	374-488
1.3	1	2-5	365-474
1.3	2	4-10	370-481
1.3	3	6-15	366-476
1.3	4	8-20	370-481

The flame propagation was captured by a CMOS camera (PHANTOM V1611) at the rate of 12000 fps. A Matlab code was deployed to get the flame radius of each image. The corresponding pressure was recorded using the pressure transducer. The flame speeds were calculated with the given radius and pressure using the given equation [3]:

$$S_u = \frac{dR_f}{dt} - \frac{\left(R_0^3 - R_f^3\right)}{3\,\gamma_u R_f^2 P}\frac{dP}{dt} \quad (1)$$

where R_f and R_0 are the flame radius and the inner chamber radius respectively and γ_u is the heat capacity ratio of the unburnt gases.

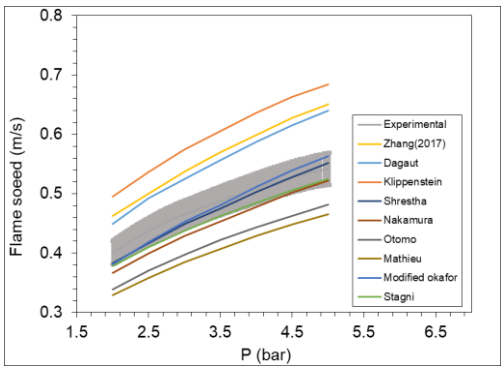

Fig. 1 Comparison of experimental flame speeds and simulated flame speeds at *φ=1.3, P_0=1 bar, T_0=296.8 K, 21% He, 49% Ar*

A relative accuracy of less than 0.5% on the flame radius was propagated leading to a maximal error lower than 5% [3]. The experimental traces are widened to account for this uncertainty.

2.3 Chemical kinetic mechanisms

A literature review was performed to select 9 of the most recent kinetic mechanisms for ammonia combustion. The simulations were performed using the premixed laminar flame speed calculator available in Chemkin-*Pro*. Sensitivity analyses were performed to identify the key reactions influencing the flame speed in these mechanisms.

3. Results and Discussion

From the sensitivity analyses, it was seen that the driving reaction is the well known $O_2 + H \leftrightharpoons O + OH$. This kind of behavior is displayed by all the chosen mechanisms at all the test conditions.

Also, it is seen that the sensitivity coefficients are pressure independent. In methane flames [3], the recombination reaction, $CH_3 + H + (M) \leftrightharpoons CH_4 + (M)$ is one of the main reactions which becomes more sensitive at higher pressure and equivalence ratio. In ammonia flames, the equivalent reaction is not highly sensitive. The dominance of the driving reaction and the absence of the equivalent recombination reaction as observed for methane flames explains the ability of all mechanisms to produce a consistent trend. All of the chosen mechanisms do not give the flame speeds within the experimental range. It is seen that some of them underestimate and some overestimate invariably irrespective of the conditions.

4. Concluding Remarks

Experiments to measure flame speeds of ammonia at elevated conditions of pressure and temperature corresponding to those encountered in spark-ignition engines and gas turbines have been performed. The test conditions range over an initial pressure of 1 to 4 bar, at an initial temperature of 300 K for three different equivalence ratios: 0.8, 1.1 and 1.3. The tests were performed in the constant volume condition which is the most efficient method to obtain data for extreme conditions. The novelty of the experimental set-up enables a complete visualization of the flame. It also permits to test for a wider range of conditions. A selected set of existing kinetic mechanisms have been evaluated. The ability of the kinetic mechanisms to capture the experimental trends is attributed to the dominance of $O_2 + H \leftrightharpoons O + OH$. Nevertheless, only 4 mechanisms among the 9 selected can predict the flame speeds within the range of the experimental values.

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