Full Length Article

Approximation of laminar flame characteristics on premixed ammonia/hydrogen/nitrogen/air mixtures at elevated temperatures and pressures

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ABSTRACT

Ammonia shows a promising potential to be a carbon-free and sustainable fuel for gas turbines and internal combustion engines. The design of such complex combustion systems using computational reactive fluid dynamics in combination with detailed reaction mechanisms is a time-consuming challenge. Combustion models are a solution to reduce the computation time, but require data of fundamental laminar flame characteristics. In order to provide such data, in this present study correlations of laminar flame speed and flame thickness for ammonia/hydrogen/nitrogen/air mixtures based on a prior selected detailed reaction mechanism were developed. A validation study for three detailed chemical schemes was conducted using experimentally obtained laminar flame speed data from literature to determine one mechanism with the best predictive capabilities. A database with 222250 data entries of laminar flame speed and flame thickness was generated by performing numerical one-dimensional simulations using the selected mechanism. From that correlations were derived. The database as well as the derived correlations contain a broad range of initial conditions: air/fuel equivalence ratio: (0.5–1.7), ammonia: (0–100) mol%, hydrogen addition: (0–60) mol%, nitrogen addition: (0–20) mol%, fresh gas temperatures: (300–1100) K and pressures: (0.1–25) MPa. For the substitution of fuel within existing burners or engines which so far are operated with methane, it is of special interest, if a defined ammonia/hydrogen/nitrogen/air mixture could be used with similar properties. For that it is shown that such a mixture composition can be found which has a similar laminar flame speed than methane, depending on the given initial conditions.

1. Introduction

The demand for a transition from fossil fuel to renewable energy sources is driven by the problematic influence of greenhouse gases as well as by limited fossil fuel resources. One option can be seen in so called electrofuels [1], being generated using renewable electricity for example in periods of excess production of wind or solar farms. In this situation the fuel has the function of a chemical energy storage system, which has much higher storage capacity than any other energy reservoir. Possible electrofuels are synthetically produced hydrocarbon or carbon free fuels. The potential advantages like storage capability, handling, safety issues and properties for the application have to be determined.

Ammonia (NH_3) is one of the promising carbon-free sustainable electrofuel like hydrogen (H_2), because only water and nitrogen are produced from complete combustion. In terms of combustions properties compared to hydrogen or hydrocarbons ammonia has a narrower flammability range, lower flame temperature and slower laminar flame speed. Due to this reasons ammonia was long time not considerate as

fuel and has been insufficiently studied. Ammonia has some benefits compared to hydrogen. For example like propane, ammonia can be stored at moderate pressures (0.8 MPa) at room temperature, so storage costs are relatively low. Furthermore production, handling, storage systems and distribution infrastructure are already established [2]. At present, mass production of ammonia via the Haber-Bosch-Process uses hydrogen from cracked fossil fuels, mainly natural gas [3]. Recent studies show that also sustainable and environmentally friendly synthesis processes are possible being based on renewable energy sources [4–6]. Hence ammonia has great potential to become one of the relevant electrofuels of the future. Also Ahlgren [7] came to the conclusion that ammonia is more suited for the transport in pipelines than hydrogen or methane. Grinberg Dana et al. [8] showed that among carbon and nitrogen based fuels, ammonia has the highest power-to-fuel-to-power index, which is the ratio of the output power from fuel's combustion to the energy required for its production and distribution. Despite the benefits of ammonia as fuel there are some drawbacks regarding the toxicity and the lower heating value. Nevertheless, there is a growing interest to use ammonia as a fuel for gas turbines and internal

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combustion engines [9–15]. First attempts to use ammonia as a fuel for conventional gas turbines were studied by Verkamp et al. [16], Newshall and Starkmann [17] and also Pratt [18] in the late 1960s. These studies showed that due to the slower reactions of ammonia the combustion system, which were designed for hydrocarbon fuels, has to be twice as large.

Verkamp et al. [16] proposed an interesting approach based on the idea to partially dissociate ammonia into hydrogen and nitrogen before injection into the combustor, as the hydrogen content promotes the combustion. He concluded that a mixture of 28% dissociated ammonia could be used as a substitute fuel in gas-turbine-combustion systems optimally sized for hydrocarbons fuels.

Recently investigation by Norihiko et al. [19] and Kurata et al. [20] demonstrate the use of ammonia/kerosene, ammonia/methane and pure ammonia in a micro gas turbine, respectively. A diffusion combustion due to flame stability reasons and selective catalytic reduction (SCR) for NOₓ reduction were employed. Valeria-Medina [21,22] studied the combustion of ammonia/hydrogen and ammonia/methane in a premixed swirl burner. He also found that a blend of 50% ammonia and 50% hydrogen has a similar laminar flame speed to that of methane. Initial attempts using pure ammonia resulted in a blow off before reaching stable conditions. Hayakawa et al. [23] achieved in model swirl burner a premixed ammonia/air combustion for equivalence ratios of 0.8, 1.0 and 1.2 at atmospheric conditions by using a bluff body.

Detailed reaction mechanisms for ammonia/hydrogen/nitrogen/air oxidation were proposed, for instance by Mathieu and Petersen [24], Tian et al. [25] and Konnov [26]. Xiao and Valera-Medina [27] compared 12 different chemical kinetic schemes for premixed ammonia/hydrogen/air combustion regarding laminar flame speed, ignition delay time and NOₓ emissions. Their results show that Mathieu's and Tian's mechanism correspond best to the experimental data. For premixed ammonia/hydrogen/air combustion Xiao et al. [28] proposed an improved detailed mechanism mostly based on Mathieu's mechanism in terms of NOx emission calculations. Further three reduced mechanisms for gas turbine conditions were developed to reduce the computation time of numerical simulations.
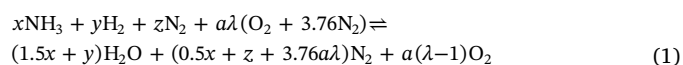
Fundamental research regarding the laminar flame speed of premixed ammonia/air and ammonia/hydrogen/air flames were conducted by Hayakawa et al. [29], Takizawa et al. [30], Jabbour and Clodic [31], Pfahl et al. [32], Ronney [33], Zakaznov et al. [34], Ichikawa et al. [35], Li et al. [36], Kumar et al. [37] and Lee et al. [38,39] mainly for atmospheric flames. It should be mentioned that in [30–34] the effect of flame stretch were not considered. Furthermore, laminar flame speed data at elevated pressures is scarce and at elevated temperatures do not exist in literature during the time of this study. Hayakawa et al. [29] and Ichikawa et. al. [35] have obtained laminar flame speeds of ammonia/air and ammonia/hydrogen/air in a constant volume combustion chamber for pressures up to 0.5 MPa, respectively. Li et al. [40] found out that the laminar flame speed of ammonia/hydrogen/air flames are quasi-linearly with the maximum mol fraction of $NH_2 + H + OH$ and multiplied by a factor k = 3758.5.

The purpose of this present study is to provide a comprehensively correlation of unstretched laminar flame speed and laminar flame thickness for premixed ammonia/hydrogen/nitrogen/air flames at elevated temperatures and pressures. Aim is that the correlation should be valid for a broad range of conditions, such that different applications like atmospheric flames, gas turbines and spark-ignition engines are contained. For that purpose three selected detailed reaction mechanisms were compared with experimentally acquired laminar flame speeds from literature. The best performed mechanism was used to generate a database containing results of one-dimensional simulations. From the obtained simulated data, laminar flame speeds and flame thicknesses were extracted and used to derivate the correlations as function of the mixture composition, temperature and pressure. The measured laminar flame speed from literature were also considered in the correlation. Further a comparison of methane and ammonia/

hydrogen/nitrogen were performed to provide ratios between ammonia and hydrogen/nitrogen to get a similar laminar flame speed to that of methane. The assumption is that such correlations can be used for the calculation of technical applications for ammonia/hydrogen/nitrogen fuel mixtures if the reaction model for turbulent premixed flames is based on laminar flame data. This is the case for common turbulent premixed flame models such as the Algebraic Flames Surface Wrinkling (AFSW) model [41–44] or Coherent Flame Model (CFM) [45]. This would allow to reduce the computation costs and to drive future development of premixed combustion systems powered by ammonia.

2. Computation setup

The overall chemical reaction for $NH_3/H_2/N_2$/air gas blends is defined by the following equations and is used to calculate the unburned mixture composition and the equivalence ratio ϕ or air/fuel equivalence ratio (in German Luftzahl) λ:

$$xNH_3 + yH_2 + zN_2 + a\lambda(O_2 + 3.76N_2) \rightleftharpoons$$
$$(1.5x + y)H_2O + (0.5x + z + 3.76a\lambda)N_2 + a(\lambda-1)O_2 \quad (1)$$
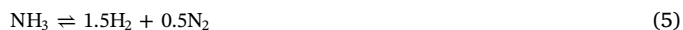
$$x + y + z = 1 \quad (2)$$

$$a = 0.75x + 0.5y \quad (3)$$

$$\lambda = \phi^{-1} \quad (4)$$

Dissociated or cracked ammonia is described by the following chemical equation:

$$NH_3 \rightleftharpoons 1.5H_2 + 0.5N_2 \quad (5)$$

Thereby the ratio of hydrogen to nitrogen is 3:1 and can be applied in the overall Eq. (4) to emulate dissociated ammonia.

The simulations of unstretched laminar flame speed and laminar flame thickness are performed with the open-source software Cantera [46] using a one-dimensional freely-propagating premixed laminar flame model with an adaptive grid refinement control and multicomponent transport. The Soret effect was neglected to reduce computation time. The following used grid refinement settings ratio = 3, slope = 0.015 and curve = 0.03 resulted in 595 grid points on average and provided a high accuracy. In order to perform automated parallel computations and all the necessary data handling a Python script incorporating the Cantera libraries [46] was written. In total, 222250 simulations were performed and required approximately 8 days of CPU time on 32 Cores (2× Dual CPU X5690 3.46 GHz, 48 GB RAM and 1x i7-5820 K 3.30 GHz, 32 GB RAM). The corresponding initial conditions are listed in Table 1. The simulation data are used to calibrate the correlations for laminar flame speed and flame thickness by using the NL2SOL Algorithm (An Adaptive Nonlinear Least-Squares Algorithm) in the software toolkit Dakota [47] afterwards. Due to the large spread in the values of laminar flame speed and flame thickness a weighting parameter, defined as the squared reciprocal of the observed value, was used. The handling of the calibration process is semi-automated through a Python script to speed up the process and to prevent errors.

The accuracy of mechanisms and final correlations are evaluated by the mean absolute percentage error $M_\emptyset$, max absolute percentage error M_{max}, min absolute percentage error M_{min} and the coefficient of

Table 1
Initial conditions for 1D simulations.

Parameter	Symbol	Unit	Range
NH₃ mole fraction	x_{NH_3}	–	0.0–1.0
H₂ mole fraction	x_{H_2}	–	0.0–0.6
N₂ mole fraction (∀ $x_{H_2} \neq 0$)	x_{N_2}	–	0.0–0.2
Air-fuel equivalence ratio	λ	–	0.5–1.7
Pressure	p	MPa	0.1–25
Unburned temperature	T_u	K	300–1100