A novel kinetic reaction mechanism for predicting laminar flame speeds of NH_3/H_2 fuel mixtures

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Abstract

The significance of ammonia (NH_3) in various human activities is evident through its extensive utilisation as a feedstock in fertilizers, industrial chemicals, and emission after-treatment systems. Ammonia's potential as a zero-carbon fuel and hydrogen (H_2) carrier has ignited scientific interest in its application as a fuel in combustion systems. However, harnessing ammonia as a fuel source for energy applications presents notable challenges due to its low flammability and the potential for high emissions. Blending NH_3 with H_2 presents a prospect for enhancing combustibility, albeit with a notable increase in NOx emissions, particularly in fuel-rich conditions. To address these challenges and underscore the underlying factors, a comprehensive understanding and analysis of the kinetic chemistry of NH_3 is imperative, particularly concerning the examination of the overall combustion characteristics of NH_3 flames employing a kinetic reaction mechanism. While numerous studies[1–16] have been conducted to analyse the gas-phase chemical kinetics of NH_3, aimed at enhancing prediction accuracy, the outcomes have been less than promising, largely due to constraints in the applicability of their refined models. These studies relied upon experimental data from existing literature [17–27], supplemented by experiments conducted within the scope of the respective studies under specific operational conditions. The limitations in kinetic model applicability can be clarified by either kinematic discrepancies resulting in deviations in the rate parameters of the reactions or mechanistic deficiencies, leading to a lack of important chemical routes. These deficiencies significantly impact the ultimate outcomes, highlighting the necessity for a more comprehensive understanding of the intricate chemistry underlying the system.

The current study focuses on tuning Arrhenius parameters of the rate constants within pre-defined uncertainty limits [28,29] using code Optima++ [30,31] to fit experimental observations from previous studies with the consideration of their associated uncertainty. The study proposes the San Diego mechanism as a baseline for improvement due to its exceptional small size (21 species and 64 reactions) and its fair overall performance demonstrated in prior study by Szanthoffer et al. [32]. Optimisation targets comprised 1311 laminar burning velocity data points in 185 data series measured in wide range of conditions, ensuring comprehensive coverage for robust model development.

The refined model showed high prediction accuracy of laminar burning velocities across wide range of operational conditions, surpassing all other kinetic models documented in the literature.

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