Optimization of ammonia combustion chemistry using NH_3 and NH_3/H_2 combustion experimental and theoretical data

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Ammonia (NH_3) stands as a pivotal player in the global shift toward carbon-free energy systems, yet its low reactivity suggests exploring blends with high-reactivity components such as hydrogen (H_2). Reliable chemical kinetic models are crucial for advancements in NH_3-based combustion technologies. Our prior work [1] comprehensively evaluated several literature models using extensive experimental data, revealing their limitations in accurately predicting combustion behavior across diverse conditions, underscoring the need for further model refinement.

To address this, we optimized ammonia combustion chemistry using an extensive amount of data from well-controlled experiments (indirect data), including laminar flame speeds, ignition delay times, and species concentrations measured in flow reactors, jet-stirred reactors, and shock tubes, as well as rate coefficient data (direct data) from measurements and theoretical calculations. The NH_3 and NH_3/H_2 combustion experimental database from [1] was expanded with recent literature measurements and our newly measured data, targeting conditions where existing models underperformed, including shock tube ignition delay times for NH_3/H_2 mixtures and flame speeds in O_2-enriched flames. A large dataset of relevant rate coefficients was also compiled from the literature. We employed our novel optimization methodology [2], which optimizes all three Arrhenius parameters of sensitive reactions in their principal component space, and has proven more accurate and robust than conventional approaches. The objective function employs a curve-matching score metric [1, 4] to quantify how closely model predictions align with experimental data in both magnitude and curve shape. Initially developed for indirect data, this approach was extended here to include direct data, ensuring an integration of all available data sources. Starting with the Zhu et al. [3] model, selected for its superior predictive performance in our prior work [1], we developed an optimized NH_3 kinetic model with a significantly improved predictive accuracy across diverse conditions. The optimization process, by modifying rate parameters, revealed globally sensitive reactions, as parameter changes reflected their influence across the dataset's conditions, thus identifying key pathways driving NH_3 combustion and prioritizing reactions for future model refinement.

References

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