

The Effects of Working Conditions on Pollutants During Ammonia Combustion

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MIT Portugal

2022 Annual Conference

Background

Fossil fuels are fundamental for our society. However, they have devastating effects on our environment and climate. Since prevailing renewable energies are intermittent in nature, large-scale energy storage solutions are necessary to store them. Recently, ammonia has been suggested as a good alternative for energy storage since it has the highest overall energy efficiency considering the complete fuel synthesis and utilization cycle.^{1,2} To use ammonia, we need to thoroughly understand its combustion process and what controls the resulting emissions at all relevant reaction conditions. The question we are interested in is **how the emissions (NO_x, unburnt NH₃) depend on reaction conditions, specifically: temperature, pressure, equivalence ratio, residence time, and water content.**

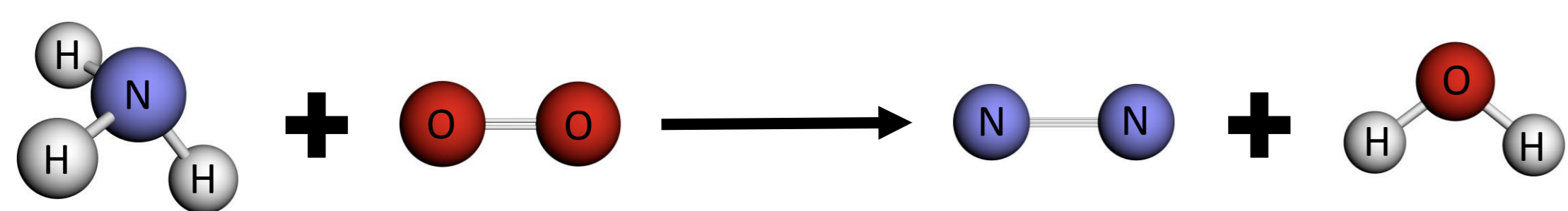
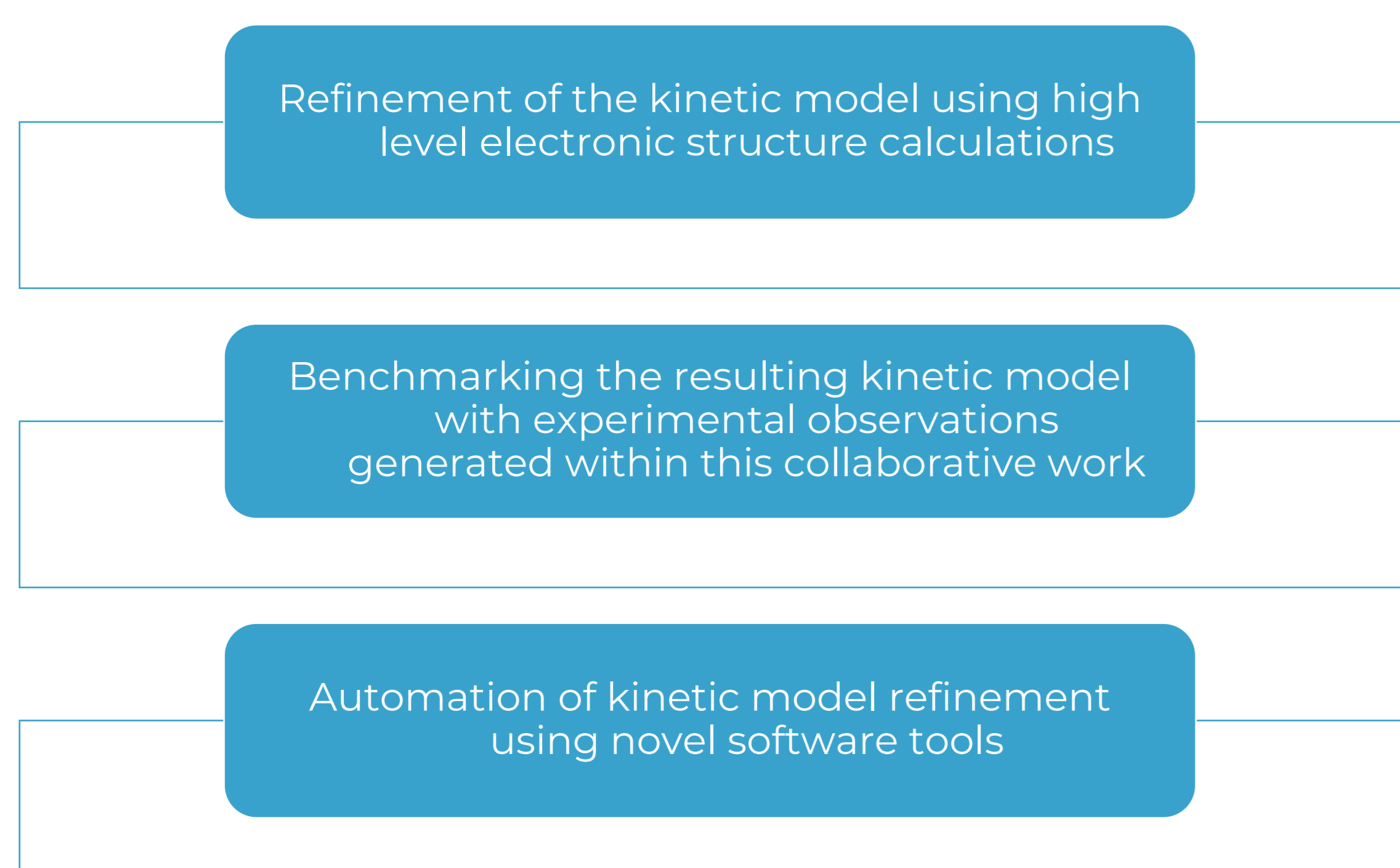


Figure 1. The complete ammonia combustion reaction.

Goals

1. Enhance our comprehension of ammonia combustion.
2. Obtain a better understanding of the effects working conditions have on pollutants.
3. Develop a state-of-the-art predictive chemical kinetic combustion model. This model will be refined using high-level electronic structure calculations and benchmarked to experimental observations generated within this collaborative work.

Challenges



Methodology

To accomplish our goals, we will do the following:

- Generate a gas-phase ammonia combustion model using an open-source software called the **Reaction Mechanism Generator (RMG)**. This tool will let us automatically generate such a model by leveraging a database of known thermochemical and kinetic parameters.³
- Update parameters for existing data using reliable quantum chemistry calculations using a software tool called **Automated Rate Calculator (ARC)**.⁴ Ultimately, we will refine the model with **The Tandem Tool (T³)** as shown in Fig. 2.⁵
- Simulation of the kinetic model with **Cantera** to compare it with experimental values as well as other kinetic models from the literature.⁶

References

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3. M. Liu et al., *Reaction Mechanism Generator v3.0: Advances in Automatic Mechanism Generation*, *J. Chem. Inf. Model.* **2021**, 61(6), 2686–2696.
4. A. Grinberg Dana et al., *ARC - Automated Rate Calculator*, version 1.1.0, <https://github.com/ReactionMechanismGenerator/ARC>, DOI: 10.5281/zenodo.3356849.
5. A. Grinberg Dana, et al., *The Tandem Tool (T³) for automated kinetic model generation and refinement*, version 0.1.0, <https://github.com/ReactionMechanismGenerator/T3>.
6. D.C. Goodwin et al., *Cantera: An Object-oriented Software Toolkit for Chemical Kinetics, Thermodynamics, and Transport Processes*, 2022, <https://www.cantera.org>, 10.5281/zenodo.6387882.
7. B. Shu et al., *Proc. Comb. Inst.* **2019**, 37(1), 205–211.

Relation of the automated tools

The three software suites and their role in the scheme of automated model generation and refinement.³⁻⁵

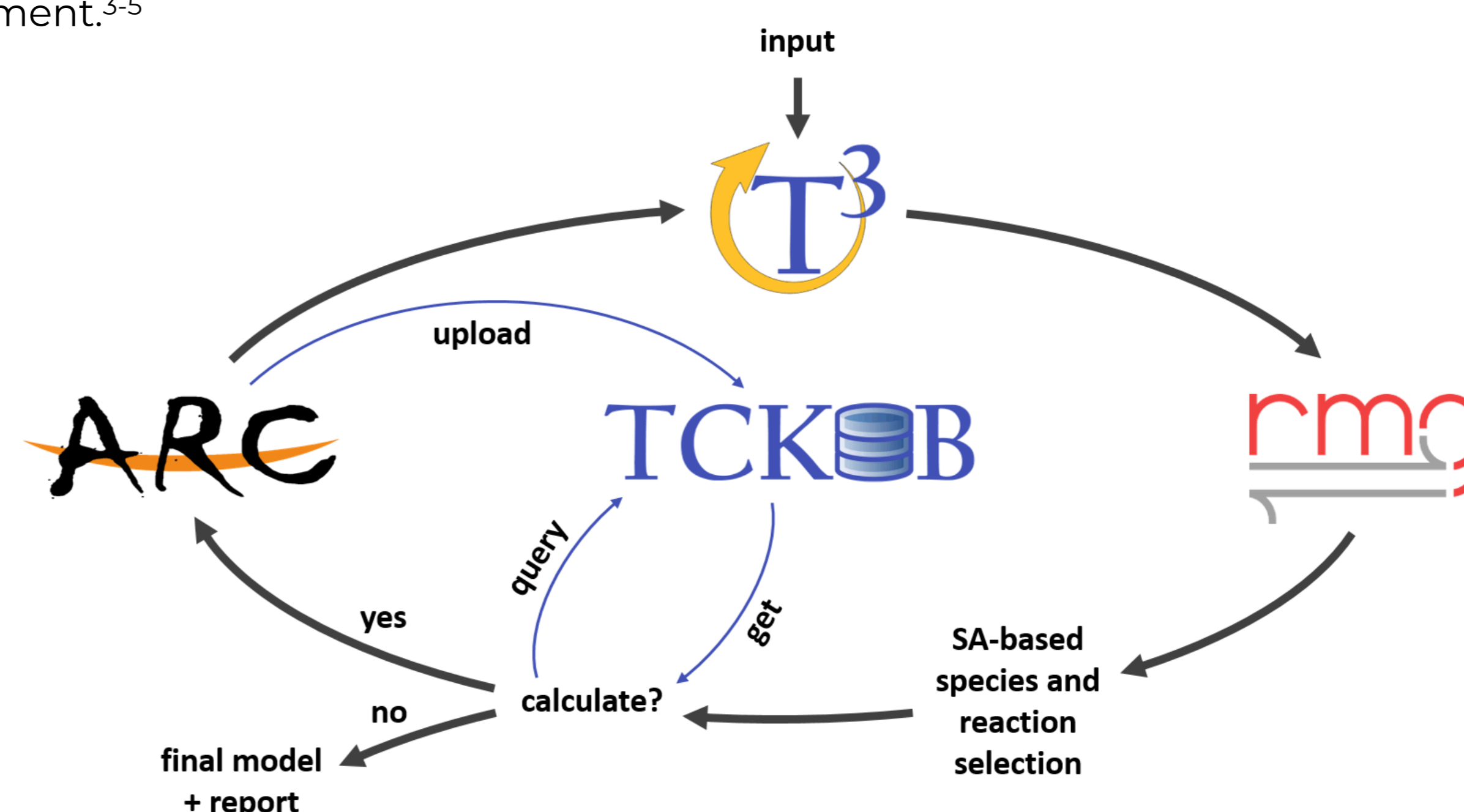


Figure 2. Scheme of model development using the automated tools that will be used in this research [3].

- RMG is used for the model generation.
- ARC is used to automatically calculate thermochemical and kinetic parameters based on electronic structure methods.
- T³ iteratively calls RMG and ARC to generate a kinetic model and refine it.

Results

Fig. 3 shows simulated and experimental values of ignition delay times of ammonia and air mixtures at 0.5, 1.0 and 2.0 equivalence ratios (Φ) at two different pressures (20 and 40 bar), and a temperature range of 1100 – 1600 K. Our model can reasonably reproduce the experimental results. Although there are still some discrepancies as pressure rises, the predicted values still capture the experimental trend.

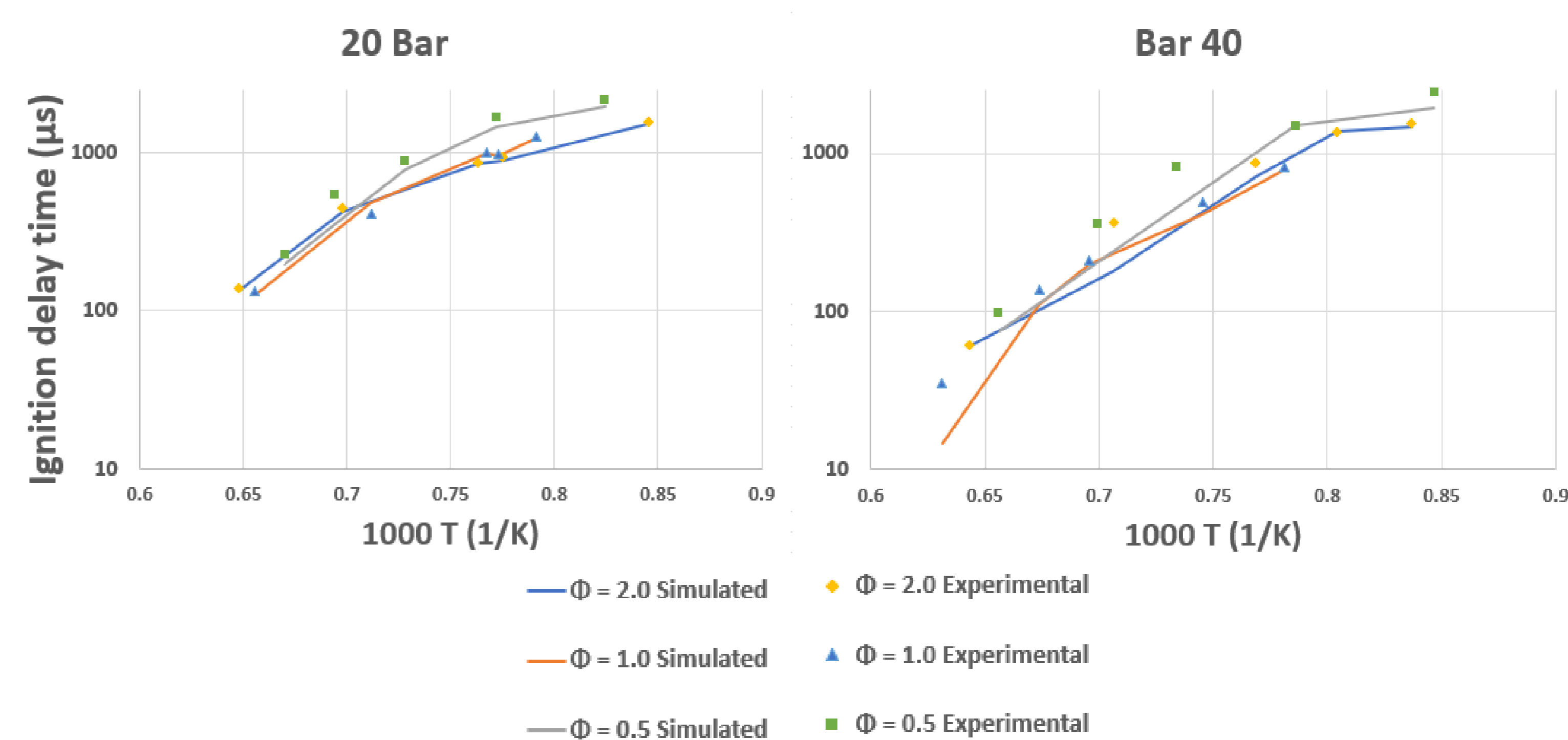


Figure 3. Effects of the equivalence ratio on the ignition delay time of NH₃/air mixtures at 20 and 40 bar. Simulation is based on our updated kinetic mechanism and the experimental results were taken from B. Shu et al.⁷

Future work

- Perform sensitivity analysis to identify most important reactions.
- Find transition state geometries and refine reaction rates.
- Compare species concentration profiles, ignition delay times, flame speed and conversion with experimental data, as well as with current literature models.

Financial support from the MIT-Portugal program 2022 is gratefully acknowledged.

