Diffusion is your friend, insights from fully coupled NH_3 plasma cracking modelling

R. Quiroz Marnef, S. Maerivoet, I. Tsonev, A. Bogaerts
PLASMANT, University of Antwerp, Antwerp, Belgium

Abstract: In this contribution, we present a fully-coupled 2D-axisymmetric model of warm NH_3 plasmas. The Navier-Stokes, heat transfer, Ohmic current continuity and chemical transport equations are solved self-consistently. The model employs a reduced thermal chemistry set to reproduce experiments of a pin-to-pin plasma reactor, therefore strengthening the claim that NH_3 plasma conversion is mainly driven by thermal processes.

1. Introduction

Reactor design improvement requires understanding of the effective chemistry and the flow dynamics, which can both be achieved by multi-dimensional models. Due to the high rate coefficients, thermal chemistry is expected to be the dominant cracking pathway at high (>2000 K) gas temperatures [1]. Furthermore, 2D-axisymmetric models have proven to be an effective tool in accurately describing the main conversion mechanisms for gas-phase chemistry in warm plasmas [2].

Here, we use a reduced chemistry set to spatially resolve the chemical processes inside an NH_3 pin-to-pin plasma cracker. We discuss the importance of diffusion and fully couple all relevant physics.

2. Method

We model an undiluted stream (5-20 NLM) of cold NH_3 injected in a pin-to-pin plasma reactor with a diameter of 16 mm. This experiment is modelled in two parts: first, the inlet of the reactor creating a swirling flow pattern is modelled with a 3D fluid dynamics simulation, and second, a 2D-axisymmetric fully coupled thermal plasma simulation, using the first simulation as input, calculates the Navier-Stokes, heat transfer, Ohmic current continuity and chemical transport equations with COMSOL Multiphysics 6.2. The conductivity is calculated based on the electron-neutral momentum transfer cross sections [3]. The chemistry set was reduced using a sensitivity analysis.

3. Results and Discussion

The model accurately describes both trends in the experimental conversion as well as absolute values. It reveals there are no significant back-reactions inside the NH_3 reactor and shows the importance of radial diffusion, which supplies unconverted gas to the hot plasma edge and transports a significant fraction of hydrogen radicals from the conducting plasma core to the unconverted gas near the plasma edge.

Figure 1 shows that all NH_3 is converted in a rather narrow temperature region between 2000-3000 K (blue curve). Furthermore, locally the cracked NH_3 is replaced by unreacted gas diffusing in from the reactor edge (red curve). The change in sign of the diffusive term indicates where NH_3 is supplied/removed by diffusion. Axial and radial convection supplies fresh cold gas from upstream the reactor. It is greater close to the reactor edge, indicating the important role of resolving the fluid flow.

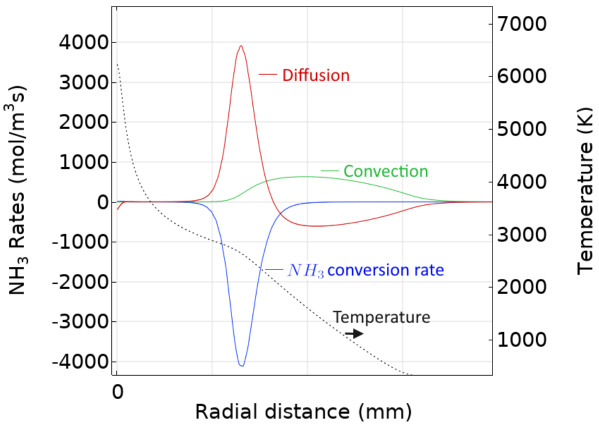

Figure 1. Contributions to the local balance of NH_3 production and loss. Chemical reactions in blue, radial diffusion in red, convection in green. The local temperature as well as the sum is also indicated. Values plotted at a cutline halfway between the electrodes for a flowrate of 15 NLM and an SEI of 35 kJ/mol.

4. Conclusion

Comparison of multidimensional modelling with experiments for NH_3 cracking demonstrates that thermal chemistry adequately describes the dominant processes. Therefore, the ionized gas mainly functions as an efficient source of heat and hydrogen radicals. As thermal chemistry-based multi-dimensional modelling can reproduce experimental results, it can be used to improve reactor design. Diffusion plays a crucial role by supplying both the unreacted gas as well as the important intermediate hydrogen radicals. The model shows that the energy cost varies remarkably little over a wide range of flow rates and deposited powers, indicating a robust operating regime.

References

[1] P. Glarborg et al., Fuel Commun. **10**, 100049 (2022).
[2] S. Maerivoet et al., Chem. Eng. J. **492**, 152006 (2024).
[3] O. Vallhagen et al, J. Plasma Phys. **86**, 475860401 (2020)
[4] B. Bayer et al., ACS Catal., **13**, 2619−2630 (2023)