Self-consistent modeling of microwave plasma in air for nitrogen fixation

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Abstract: In this work, we developed a 2D-axisymmetric fully coupled thermal microwave air plasma model. By coupling the microwave field heating to the reactive flow, the model is the first fully self-consistent model for microwave air plasma. Our model calculations align perfectly with experimental NO_x measurements at 0.65 bar, 20 slm and 400–700 W.

1. Introduction

Plasma-driven nitrogen fixation is a promising sustainable alternative for the Haber–Bosch process due to its compatibility with renewable sources **[1]**. A recent techno-economic study by Rouwenhorst *et al.* showed that plasma-based fertilizer production may be industrially competitive if the energy cost (EC) can reach 1–1.5 MJ/mol **[2]**.

Microwave plasma is a promising candidate due to its well-known advantages, including efficient power coupling, scalability and absence of impurities originating from the electrodes. Kelly *et al.* achieved the lowest EC of 2 MJ/mol for microwave plasma-based NO_x production at atmospheric pressure **[3]**.

Despite these encouraging results, comprehensive multidimensional models of microwave air plasma at atmospheric pressure remain scarce **[4, 5]**, hindering further optimization of the technology. Moreover, these models typically estimate the power density from emission spectra, which is associated with significant uncertainty, and which limits the predictive character of the models. We present the first fully self-consistent model for microwave air plasma by coupling the microwave field heating to the reactive flow.

2. Method

The 2D-axisymmetric microwave air plasma model solves the Navier–Stokes equations, heat transfer, reactive species transport and electromagnetic wave equations. The physics are solved within COMSOL Multiphysics 6.2, using a fully-coupled solver. A small thermal chemistry set of 12 reversible reactions is implemented, including N_2/O_2/NO dissociation, the Zeldovich mechanism and NO oxidation.

3. Results and Discussion

Figure 1 (a) shows the calculated temperature profile at 440 W, where the peak temperature of 5400 K agrees with the experimentally measured value at the same conditions **[5]**. Figure 1 (b) illustrates the NO molar fraction, and it is clear that NO is mainly formed at the edges of the plasma. Here, NO is quenched by radial diffusion and subsequently transported to the outflow by axial convection.

Figure 2 presents the comparison of outflow NO_x concentration between model and experiment, wherein the model calculations perfectly align with the experimental results **[5]** within the power range of 400–700 W.

4. Conclusions

We developed the first fully self-consisted microwave air plasma model by coupling the microwave field heating to the reactive flow, obtaining excellent agreement with experiment **[5]**. This agreement illustrates the predictive capabilities of the model, making it a promising tool for process optimization and reactor engineering.

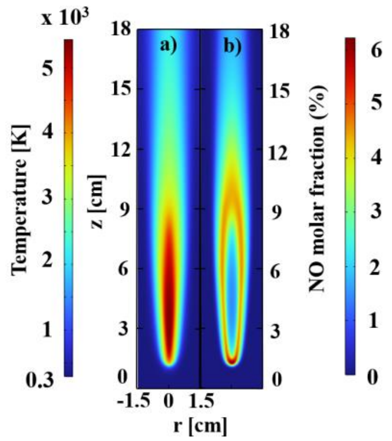

Figure 1. Calculated temperature (a) and NO molar fraction (b) profiles at a power of 440 W.

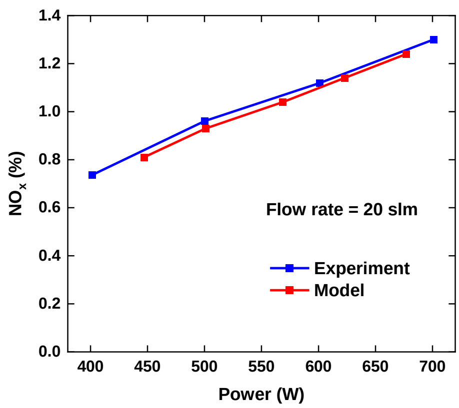

Figure 2. Comparison between model and experiment **[5]** for NO_x concentration, measured at a pressure of 0.65 bar, flow rate of 20 slm and power of 400–700 W.

References

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