2D Raman temperature imaging in a microwave air plasma

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Abstract: Microwave air plasma is studied in the context of electrification of nitrogen fixation. We introduce in situ temperature measurements by 2D Raman scattering, which infers vibrational temperature. The technique reveals core temperatures at 5000 K, far beyond the thermal optimum for NO_x production, and broadening of temperature distributions with higher power in constant specific energy input cases.

1. Introduction

Plasma technology offers sustainable industrial solution for nitrogen activation to meet the rising demand for agricultural fertilizers, while mitigating global greenhouse gas (GHG) emissions caused by the production. Industrial plasma-based nitrogen fixation was first realized with the Birkeland-Eyde (B-E) process for NO_x synthesis. The plasma approach may enable decentralized fertilizer production, with air as the feed gas and compatibility with intermittent energy sources. Understanding temperature and plasma chemistry is critical for obtaining key scientific insights to optimize the process [1, 2]. In this work, we present a two-dimensional (2D) Raman scattering technique [3] enabling in situ temperature and N_2 density imaging.

2. Methods

The experiments were conducted using microwave air discharge at 300 mbar, with a swirl gas injector. A 532 nm Nd:YAG laser sheet was used, focused at the center of the microwave plasma. Bandpass filters were used to capture the vibrational Stokes and anti-Stokes Raman bands of N_2 and their ratio was used to infer the vibrational temperature from a calculated calibration curve [3]. Spectrally resolved rotational and vibrational Raman measurements validated the technique as well as confirmed equilibrium between vibrational and rotational temperatures. Hence, we will denote both as “gas” temperature in this work.

3. Results and Discussion

The 2D Raman results for the gas temperature and density of N_2 in a microwave air plasma are shown in Figure 1. In this figure, the specific energy input (SEI) remains constant at 54 kJ/mol, between two measurements. The broader and more uniform temperature distribution achieved in 800 W, 20 SLM is in line with plasma broadening at higher power applied in combination with faster radial transport at higher temperature. Besides, at higher power, NOx production is less efficient, which is in line with the larger region over overheating compared to the ~3000 K optimum for thermal NO production. The steeper edge temperature gradient may be influenced by stronger vorticity at the higher input flow.

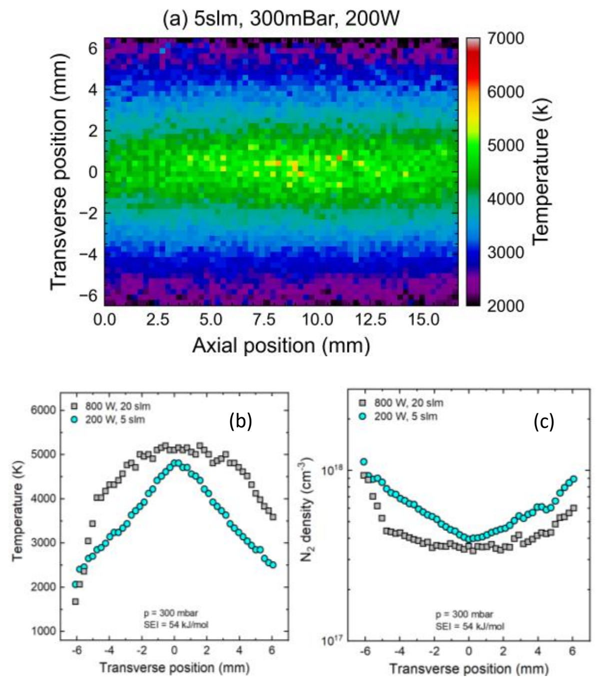

Fig. 1. a) Measured 2D image of radial cross section of reactor, showing vibrational temperature in air plasma of 200 W. b) Vibrational temperature, and c) N_2 density measured in air plasma at 800 W and 200 W, SEI = 54 kJ/mol

4. Conclusion

We chose two conditions in which SEI was kept constant by simultaneous variation of power and flow. The in situ measurements demonstrate that plasma and gas interaction is very much different due to changes in peak temperature, temperature profile and radial transport. Thus, SEI seems not to be a valuable dimensionless parameter for reactor scaling. Furthermore, measured temperatures of 4500 - 5000 K were far in excess of the B-E process gas temperature optimum of ~3000 K for NO plasma synthesis.

References

[1] A. Fridman, Plasma chemistry, Cambridge University Press (2008)
[2] N. Gatti et al., Plasma Sources Science and Technology 27, 5 (2018)
[3] Del Cont-Bernard et al., Journal of Quantitative Spectroscopy and Radiative Transfer, 328, 109145 (2024)