

LIF Study of NH and OH Radicals in a Nanosecond Pulsed Discharge in a N₂:H₂O Mixture at Atmospheric Pressure

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Abstract: In this study, we present time- and space-resolved measurements of NH and OH radicals at atmospheric pressure in a nanosecond-pulsed plasma generated in humidified N₂, using laser-induced fluorescence spectroscopy. The findings provide valuable insights into NH kinetics, a key precursor for NH₃ formation, by analyzing the roles of OH and gas temperature.

1. Introduction

The nitrogen oxidation process ((H)NO_x synthesis) is well-studied in the contexts of nitrogen fixation and combustion. In contrast, nitrogen reduction for NH₃ synthesis in electrical discharges has received less attention. Direct NH₃ synthesis requires an H-containing feedstock, with H₂O being the most promising due to both its abundance and its sustainability. However, the kinetics of nitrogen reduction with water in plasma remain unexplored[1]. This study employed laser-induced fluorescence spectroscopy (LIF) to observe the time-resolved 2D distribution of NH and OH radicals in a nanosecond pulsed plasma.

2. Methods

A nanosecond-pulsed discharge (10 ns FWHM duration, 10 Hz repetition rate) was used in a dielectric barrier discharge (DBD) configuration, as shown in Figure 1a. Experiments were conducted at atmospheric pressure in a humidified N₂ flow with absolute humidity (AH) ranging from 0.01% to 2.21%.

Gas heating kinetics were monitored via Rayleigh scattering. The LIF arrangement for OH radical measurements is described elsewhere[2]. NH was detected via the NH(X³Σ⁻–A³Π, v'–v'' = 1–2) transition, exciting the P₁(4) rotational level with a 306.58 nm laser. Fluorescence was captured using an ICCD camera with a 337 ± 10 nm band-pass filter. Emission overlap from N₂ SPS was minimized by starting the measurements 1 μs after plasma pulse initiation and capturing a background image with a detuned laser at fixed laser energy.

3. Results and Discussion

Figure 1a shows 2D NH and OH LIF profiles measured at an absolute humidity of 0.22% at the delay time of 10 μs after the plasma pulse onset. Figure 1b displays the intensity profiles along the region of interest (ROI in Figure 1a) and the corresponding temperature profile. The NH signal exhibits a ring-shaped profile, and its intensity drop corresponds to the peak OH signal and temperature. The reaction chemistry analysis indicates that OH +

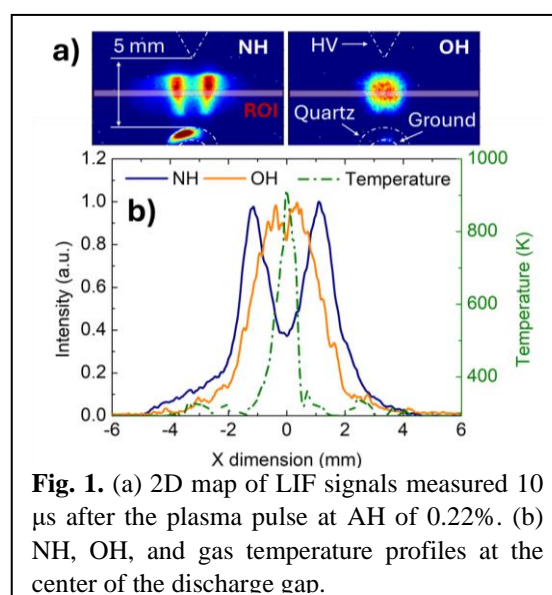


Fig. 1. (a) 2D map of LIF signals measured 10 μs after the plasma pulse at AH of 0.22%. (b) NH, OH, and gas temperature profiles at the center of the discharge gap.

N → NH + O is the main pathway for NH formation. Meanwhile, the drop in NH intensity in the center of the plasma region is explained by temperature-promoted NH removal through competitive processes, resulting in the formation of HNO, H₂O, and NH₂.

4. Conclusion

The investigation of NH radical kinetics through LIF experiments reveals that OH radicals play a crucial role in both the formation and loss processes. The gas temperature primarily determines the selectivity between further hydrogenation (i.e., NH → NH₂ → NH₃) and NH loss.

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References

- [1] M. Gromov et al., RSC Sustain., (2025). [2] M. Gromov et al., Plasma Sources Sci. Technol., **30**, 065024 (2021)