Formation of O and H radicals in an atmospheric-pressure nanosecond pulsed discharge in helium with water vapour admixtures

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1 Introduction

Plasma-produced O-, H- and N-species that have a high oxidative power and play major roles in biological functions are extensively studied for applications. In particular, plasmas containing water vapour are studied for the production of reactive species such as OH, H_2O_2, HO_2 [1]. Even though the plasma chemistry is complex and not fully understood, it is clear that atomic oxygen and hydrogen radicals play a crucial role in these plasmas.

The aim of this work was to investigate the kinetics of reactive O and H radicals in a fast pulsed discharge (rise rate ~80 V/ns) generated at atmospheric pressure in humid Helium (up to 0.25% H_2O). O and H densities are obtained experimentally by ps-TALIF (Two-photon Absorption Laser Induced Fluorescence) and are combined with 1D fluid modelling to investigate the reaction mechanisms for O and H production.

2 Methodology

The plasma has a pin-to-pin electrode geometry with a gap of 2.2 mm. A discharge in Helium with a water vapour content of 0.1% – 0.25% is created through a positive high-voltage nanosecond pulse to one of the pins. The pulse has a voltage of about 2 kV, 35 ns rise time (10-90%) and 90 ns duration FWHM. The energy dissipated in the discharge is 90 μJ and the repetition rate is 5 kHz.

TALIF is used to measure the densities of O and H radicals. Details of this method can be found in [2]. The 1D plasma fluid model used is based on a system of differential equations: continuity of the flux density of electrons, electron energy, ions, and neutral species, and Poisson's equation. Full details can be found in [2].

3 Results

The results of the TALIF experiments and the 1D fluid modelling are presented in figures 1 and 2. Figure 1 shows the temporal evolution of the O density from both the experiment and the model for H_2O admixtures of 0.1% and 0.25%. Figure 2 shows the equivalent H densities.

The model and experiment have a reasonable agreement, within a factor of 2 for both densities. From both figures it is clear that the main production of O and H occurs after the voltage pulse has finished. For 0.1% water, the O density builds up over ~1-2 ms to a value of approximately 2×10^{16} cm^{-3}. From 10-100 ms, the O density decays again. For 0.25% water content the O density is enhanced by roughly 40%. In the experiments, the peak in O density occurs earlier, at about 0.3 ms. This is not reproduced in the modelling.

The situation for H is similar, for both water admixtures there is a rapid increase in H density in the early pulse afterglow (<1 ms). Densities of the order of $0.2–1.0 \times 10^{16}$ cm^{-3} are observed, with the densities for 0.25% water being 50-100% higher than 0.1% water.

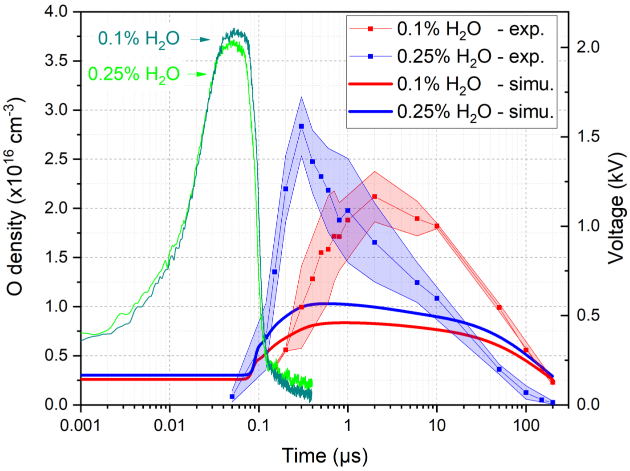

Figure 1: Temporal evolution of O density for 0.1% and 0.25% water admixture. Both modelling and experimental results are presented [2].

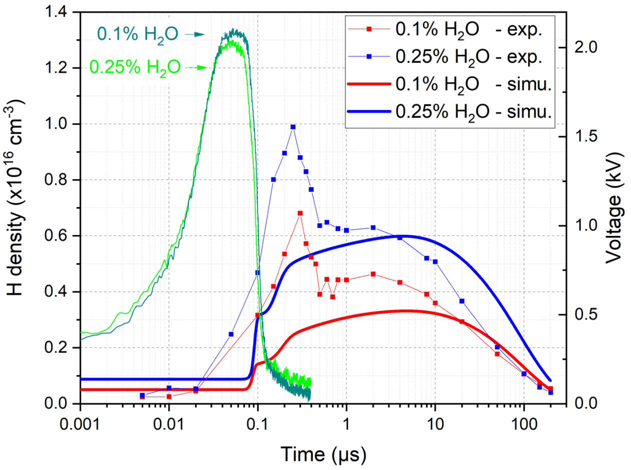

Figure 1: Temporal evolution of H density for 0.1% and 0.25% water admixture. Both modelling and experimental results are presented [2].

4 Discussion and conclusion

The delay of the production of O and H relative to the voltage pulse is surprising since it is often assumed that the dominant production mechanism is direct electron-impact dissociation. Analysis of the reaction pathways from the modelling show that there is a distinct change in reaction mechanisms during the discharge cycle. During the voltage pulse O is predominantly produced through electron-impact dissociation of O_2. For H, the dominant mechanism is electron-impact dissociation of H_2O. However, most of the O and H radicals are produced in the early afterglow (up to 1 µs) through recombination processes of O_2^+ and H_2^+. It can therefore be concluded that not only the discharge itself, but also the (early) afterglow of pulsed discharges need to be considered when studying O and H-based plasma chemistries.

Acknowledgments

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References

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