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Measure of N_2O density in a high pressure homogeneous air plasma using laser absorption spectroscopy in the infrared

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High pressure (near atmospheric) non-equilibrium plasmas in N_2/O_2 produced by different types of electrical discharges are widely studied for a variety of applications [1]. In such plasmas are created various nitrogen oxides (NO, NO_2, N_2O_5, N_2O) and the ozone molecule. Amongst these compounds, the creation-loss balance of nitrous oxide involves exited states of atomic and molecular oxygen and nitrogen [2], in particular metastables $N_2(A^3\Sigma^+_u)$ and $O(^1D)$ which play important role in the removal of hydrocarbons and VOCs. Therefore the measure of the N_2O density should help in the understanding of the excited states kinetics, if measurements can be compared to predictions of a self-consistent modeling of the discharge and plasma reactivity. This is the case for the photo-triggered discharge [3]. Formerly designed for high-power laser technology, this type of pre-ionized discharge has been later applied to gas mixtures relevant for removal of atmospheric pollutants [4-7]. It allows to create a transient homogeneous plasma (without streamer development) in various types of gas mixtures.

The photo-triggered reactor used in the present experiment has been already described [3, 4, 7]. The discharge duration is 60 ns and the plasma volume is 50 cm^3, i.e. 1 cm electrode gap and a length equal to 50 cm. A gas compressor is used to produce a gas flow through the gap. The discharge frequency is chosen such that the whole reactor volume, 500 cm^3, is renewed between two discharges. The volume of the experimental device, which corresponds to the total volume of the gas mixture studied, is V_D = 4 litres. The pressure of the studied N_2/O_2 mixtures has been fixed to 460 mbar. The experiment has been performed at ambient temperature. We measured the density of N_2O by infrared absorption spectroscopy using a Quantum Cascade Laser (QCL, Q-MACS from Neoplas Control GmbH) working in the interpulse mode (no time resolution). The wavenumber of this QCL can be scanned between 1274 and 1278 cm^{-1}, and four important optical lines of the nitrous oxide molecule [8] can be used to measure its density, [N_2O].

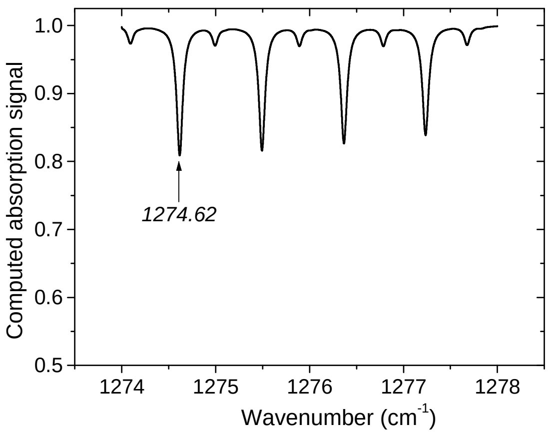

Fig. 1: Full QCL-wavenumber range absorption spectrum for 200 ppm of N_2O in air at 460 mbar total pressure and 20°C (Molspec simulation), with indication of the line used for the experiment on the photo-triggered reactor (absorption path equal to 73 cm).

The laser beam was collimated to a diameter of about 5 mm and sent through the electrode gap along the reactor length, 73 cm. The measured absorption signal was compared to a computed one making use of the Molspec software [9]. In figure 1 is displayed an example of computed spectrum over the whole range of accessible wavenumber values, showing the optical transition (centered at 1274.62 cm^{-1}, i.e. 7.845 µm) used to measure [N_2O].

In practice, the density was measured as a function of the number of discharges performed in the device volume V_D. A linear increase of [N_2O] was obtained for each chosen values of the experimental parameters (oxygen percentage, from 2 % up to 20 %, and electrical energy deposited in the plasma per current pulse, 3.5 and 4.6 J). The figure 2 gives the concentration values (ratio between [N_2O] and the total molecule density) obtained for 20 % of oxygen and an energy equal to 4.6 J (92 J/l). Thereafter the N_2O density created in the discharge volume per current pulse was straightforwardly deduced from the slope of the linear fit of experimental points multiplied by the ratio of the device volume over the discharge one. In figure 3 is plotted the so-measured molecule concentration as a function of the oxygen percentage, for the two chosen energy values.

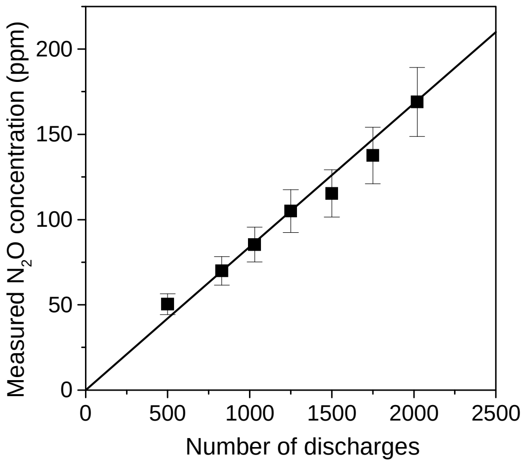

Fig. 2 : N_2O produced in the experimental device volume for 20 % of oxygen and 4.6 J of energy.

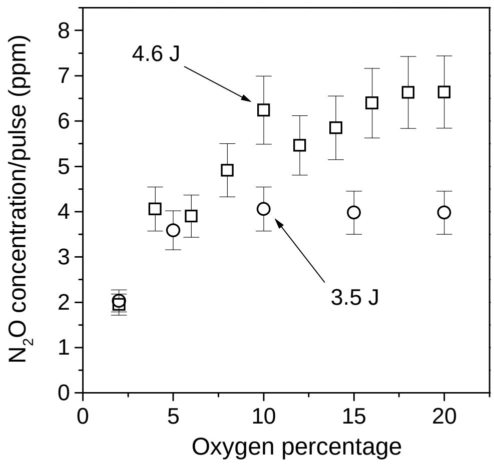

Fig. 3 : N_2O produced in the discharge volume per current pulse, for two deposited energy values.

Experimental results will be now compared to the predictions of a self-consistent 0D discharge model for N_2/O_2 mixtures [4] and the main mechanisms for N_2O productions and losses will be discussed.

References

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