O ATOM DENSITY MEASUREMENTS IN A CAPILLARY NANOSECOND DISCHARGE: MEASUREMENT TECHNIQUES AND KINETIC MODELING

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ABSTRACT

A nanosecond repetitively pulsed discharge in a quartz capillary filled with flowing synthetic air was investigated as a benchmark to address the mechanism of fast gas heating for conditions of up to complete dissociation of O_2 and heating of a few thousand K occurring during the near afterglow phase. Gas temperature, energy deposition, and O atom concentrations were measured with respect to time. O atom results are compared to the output from a detailed 0D kinetic model describing the excited species chemistry. The high oxygen dissociation degree enables investigation of the key role played by O atoms in fast gas heating of nanosecond discharge plasmas at high specific energy loading.

1. INTRODUCTION

Nanosecond repetitively pulsed discharge plasmas have been intensely studied for applications ranging from plasma assisted combustion[1] to aerodynamic flow control at high Mach number[2]. Such discharges require a low average electrical power to be operated, and demonstrate a high energy efficiency for the production of radicals and excited species[3].

When a sufficient percentage of the deposited energy is spent to the excitation of electronically excited states, these excited states can transfer a part of their energy to the gas thermal energy, through gas-phase chemical reactions occurring at a very high rate[4]. This phenomenon, called fast gas heating, can for example create shock waves that can be used in aerodynamic flow control[5].

In this paper, a detailed experimental study of fast gas heating in synthetic air at a pressure of 27 mbar is proposed, and the obtained experimental results are interpreted with the help of a detailed 0D kinetic model.

The results include deposited energy, gas temperature, and O atom density, measured by two different techniques: nanosecond actinometry with argon atoms, and TALIF of the O(3p ^{3}P) excited state of oxygen. Both of these results on O atoms are then compared to the prediction of the detailed kinetic model that is described in detail in [4], with additions described in [6].

2. EXPERIMENTAL SETUP

A schematic of the discharge tube assembly used in this work is shown in Fig. 1.

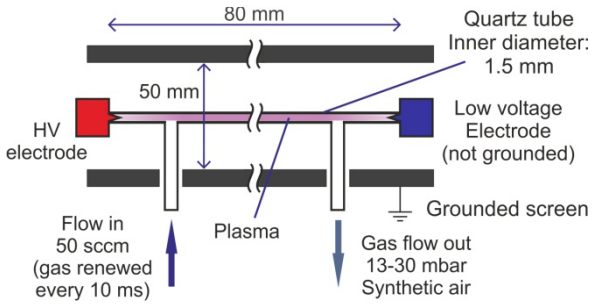


Fig. 1 Discharge tube assembly

The discharge is initiated in a quartz capillary tube, 1.5 mm in diameter and 80 mm in length,

terminated by two gold-covered pin-shaped electrodes, called the high voltage (HV) and low voltage (LV) electrodes. The capillary is filled with flowing synthetic air at a pressure of 27 mbar with a 50 sccm rate, insuring efficient gas renewal between pulses. The discharge takes the form of a fast ionization wave (FIW), followed by an energy deposition phase, during which a spatially uniform reduced electric field (E/N ≈150-300 Td, 1 Td = 10^{-17} V-cm^2) coexists with a high electric current of about 70-100 A.

The voltage pulse (29 ns FWHM, 9.8 kV amplitude, and 4 ns rise time) is supplied to the HV electrode by a FID FPG 10-MKS20 high voltage generator through a 25 m coaxial RG213 cable, called the HV cable. Three pulses separated by 250 ns are incident on the capillary tube due to successive reflections between the generator and the discharge assembly, causing three breakdowns.

The LV electrode is connected to a 100 or 200 m long RG213 cable, called the delay cable. Due to reflections from that delay cable, two additional low-energy breakdowns are created at 1 and 2 µs, that are used for optical emission spectroscopy (OES).

Incident, reflected and transmitted voltage and current pulses are registered with the help of two back current shunts (BCS), installed 12.5 m before and after the discharge assembly, in the HV and delay cables, respectively. The local electric field is obtained from potential measurements, carried out with a capacitive probe. Gas temperature is measured through rotational OES of the second positive system of molecular nitrogen, namely the $N_2(C^3\Pi_u, v = 0) \rightarrow N_2(B^3\Pi_g, v = 1)$ transition.

3. ACTINOMETRY

To measure O atom density during the first breakdown, actinometry of the O(3p ^{3}P) excited state of atomic oxygen was used, through a small (5.6%) admixture of argon atoms to the gas flow. Radiation from the $Ar(2p_1) \rightarrow Ar(1s_2)$ transition, at 750.4 nm and the $O(3p^3P) \rightarrow O(3s^3S)$ transition at 844.6 nm were registered during the breakdown, and compared. The principle of the measurement is that these two transitions are populated mainly by electron impact from the ground state, with excitation cross sections that behave similarly with electron energy [7]. This way, Ar atoms, whose density is known, can be used as surrogates for O atoms.

However, it was found in [8, 6] that secondary population processes, especially for the O(3p ^{3}P) excited state, are not negligible compared to the direct electron impact population mechanism, hence they must be taken into account, and their contribution to the OES signal subtracted. These processes are dissociative excitation, and stepwise excitation from the O(^{1}S) and O(^{1}D) metastable states. The detailed calculations from [6] give:

$$[\mathrm{O}] = A_1 - A_2 - A_3 - A_4 \quad (1)$$

$$A_1 = \frac{k_e^{\mathrm{Ar}}[\mathrm{Ar}]}{k_e^{\mathrm{O}}} \frac{\tau_{O^*}}{\tau_{Ar^*}} \frac{c_{750}}{c_{844}} \frac{f(I_{\mathrm{O}})}{f(I_{\mathrm{Ar}})} \quad (2)$$

$$f(I_{\mathrm{X}}) = \frac{dI_{\mathrm{X}}}{dt} + \frac{I_{\mathrm{X}}}{\tau_{\mathrm{X}}} + I_{\mathrm{X}} \sum_i k_{i,Q}^{\mathrm{X}} [\mathrm{Q}_i] \quad (3)$$

$$A_2 = k_{de}^{\mathrm{O_2}}[\mathrm{O_2}]/k_e^{\mathrm{O}} \quad (4)$$

$$A_3 = k_e^{\mathrm{O(^1S)}}[\mathrm{O(^1S)}]/k_e^{\mathrm{O}} \quad (5)$$

$$A_4 = k_e^{\mathrm{O(^1D)}}[\mathrm{O(^1D)}]/k_e^{\mathrm{O}} \quad (5)$$

where [O] is the absolute O atom density, k_e^{X} is the direct electron impact excitation rate for species X, τ_{X} is the radiation decay lifetime of the excited state of species X and I_{X} is the photon count of the light collected from that state; c_{XXX} is the optical collection efficiency factor for wavelength XXX, $k_{i,Q}^{\mathrm{X}}$ is the quenching coefficient of species X on species Q_i, and $k_{de}^{\mathrm{O_2}}$ is the dissociative excitation rate for molecular oxygen.

The relevant excitation constants, as well as the concentrations of metastable oxygen atoms were calculated with the help of a kinetic model [8] that was solved through the ZDPlasKin code [9].

4. TALIF

The O atom density 200-2000 ns after the first breakdown was measured with the two-photon absorption laser induced fluorescence (TALIF) technique. The principle of the TALIF technique is described in detail in our previous paper [10].

A schematic of the TALIF experimental setup used in this work is in Fig. 2. A tripled Nd:YAG laser (355 nm) was used to pump a Coumarin 2 dye laser (peak efficiency at 450 nm) which was then doubled to obtain wavelengths around 225.58 nm (for O atoms) and 224.24 nm (for Xe atoms). The beam energy was measured by an amplified energy meter, with sensitivity 1100 V/J, installed after the discharge tube, and the energy was stabilized to the desired value with the help of a variable attenuator.

An optical fiber placed above the discharge was used to collect the fluorescence light at 844.6 nm (for O atoms) and 834.68 nm (for Xe atoms) through a narrow bandpass filter, and was detected by a Hamamatsu R3896 red-sensitive photomultiplier tube (PMT) connected to the oscilloscope.

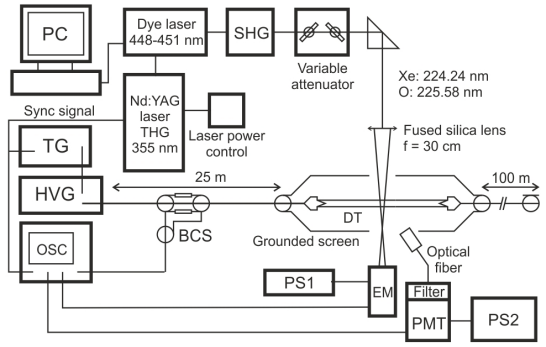


Fig. 2 TALIF experimental setup. SHG/THG: Second/Third Harmonic Generation; TG: Triggering generator; HVG: High Voltage Generator; OSC: Oscilloscope; BCS: Back Current Shunt; DT: Discharge tube; EM: Energy meter; PS: Power supply; PMT: Photomultiplier;

The delay between the laser and the discharge was adjusted from 200 to 2200 ns, allowing five measurement points. The uncertainty on the delay is ±20 ns.

5. RESULTS AND DISCUSSION

The integral energy deposited in the discharge, measured from BCS measurements, is in Fig. 3. It can be seen that the first two pulses deposit a significant amount of energy in the plasma, amounting to more than 1 eV/molecule.

The temperature measurement for 27 mbar pressure is shown in Fig. 4.

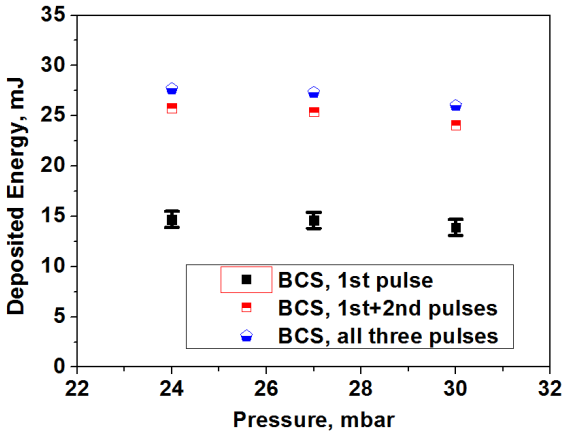


Fig. 3: Energy deposited in the discharge by the three pulses

Heating up to 2500 K is observed, that has to be taken into account in TALIF measurements because of a possible temperature dependence of the quenching parameters. To date, to the best of our knowledge, no temperature-dependent data exists in the literature for the quenching of O atoms on N_2 and O_2

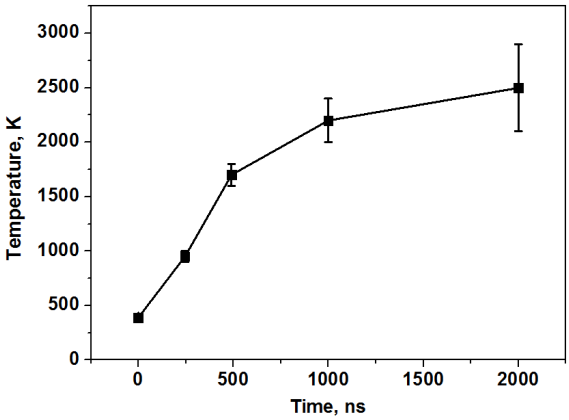
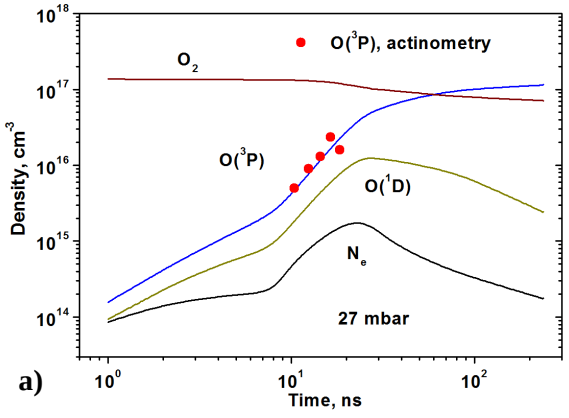


Fig. 4: measured gas temperature with time

The final result for O atom concentrations from actinometry and TALIF, are in Fig. 5.a and b, and are compared to the detailed 0D kinetic model.



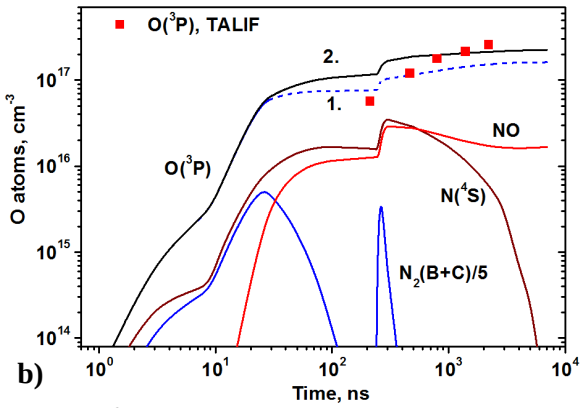


Figure 5. $O(^3P)$ atom density, and other main species, calculated by the 0D model, compared to experimental data. a) first pulse and early post-discharge, with actinometry measurements; b) 2 µs time scale, with TALIF measurements. In b), curve 1 includes reactions of quenching of electronically excited N_2 molecules on O atoms leading to NO formation, while curve 2 does not.

Early actinometric data is properly reproduced by the model; this fact gives confidence in the actinometry technique in nanosecond discharges for O atom measurement. The situation for TALIF measurements is however more complex: including reactions of quenching of N_2(A,B,a1,C) on O atoms leading to formation of NO correlates well especially with the points before 1 µs, while removing them from the scheme leads to an overestimation of the O atom density overall. It should be noted that quenching coefficients were taken here as scaling with the square root of the gas temperature (constant cross section assumption), which leads to a factor of 2 increase compared to the case with quenching coefficients that are constant with temperature. If reality is between these two possibilities, then the correlation between model and experiment could be even better.

These results indicate the key role played by reactions of quenching by O atoms in high specific energy loaded nanosecond plasmas, which has to be investigated further in the frame of studies concerning fast gas heating and plasma assisted combustion.

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