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Dynamics of reactive oxygen species produced by the COST microplasma jet

Sascha Chur^{1,*} , Robin Minke¹ , Youfan He² , Máté Vass^{3,4} , Thomas Mussenbrock³,
Ralf Peter Brinkmann² , Efe Kemaneci² , Lars Schücke³,
Volker Schulz-von der Gathen⁵ , Andrew R Gibson^{6,7} , Marc Böke⁵ 
and Judith Golda¹ 

¹ Plasma Interface Physics, Ruhr University Bochum, Bochum, Germany

² Chair of Theoretical Electrical Engineering, Faculty of Electrical Engineering and Information Technology, Ruhr University Bochum, Bochum, Germany

³ Chair of Applied Electrodynamics and Plasma Technology, Faculty of Electrical Engineering and Information Technology, Ruhr University Bochum, Bochum, Germany

⁴ Institute for Solid State Physics and Optics, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary

⁵ Experimental Physics II: Physics of Reactive Plasmas, Ruhr University Bochum, Bochum, Germany

⁶ Research Group for Biomedical Plasma Technology, Faculty of Electrical Engineering and Information Technology, Ruhr-University Bochum, Bochum, Germany

⁷ York Plasma Institute, School of Physics, Engineering and Technology, University of York, York, United Kingdom

E-mail: sascha.chur@rub.de

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Abstract

This study is focused on measuring the densities of the excited molecular oxygen species, $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$, produced in a COST atmospheric pressure plasma jet using a helium–oxygen mixture. Knowledge of the ozone density is critical for measurements because of its high quenching rate of these species. Additionally, $O_2(a^1\Delta_g)$ is difficult to measure due to its low emission intensity and sensitivity to background interference in the plasma region. Therefore, a flow cell was used to enhance signal detection in the effluent region. To validate the measurements and improve understanding of reaction mechanisms, results were compared with two simulation models: a pseudo-one-dimensional plug flow simulation and a 2D fluid simulation. The plug flow simulation provided an effective means for estimating species densities, with a fast computation time. The 2D simulation offered a more realistic description of the flow dynamics, which proved critical to correctly describe the experimental trends. However, it requires long computation times to reach an equilibrium state in the flow cell. Otherwise, it leads to discrepancies to the experimental data. Further discrepancies arose from an overestimation of the ozone density from the models, as validated from the $O_2(b^1\Sigma_g^+)$

* Author to whom any correspondence should be addressed.



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density measurements. Optimizing the reaction rate coefficients for the effluent region might improve the agreement with the experimental results. Despite these limitations, both simulations aligned reasonably well with experimental data, showcasing the well validated plasma chemistry of the models, even for complicated effluent geometries.

Keywords: ozone, $O_2(b^1\Sigma_g^+)$, $O_2(a^1\Delta_g)$, COST reference micro plasma jet

1. Introduction

Micro atmospheric pressure plasma jets (μ APPJs)[1–6] have garnered significant interest due to their wide-ranging applications, including surface treatment [7, 8], etching [9], and deposition [6, 10]. These plasma sources offer several advantages over traditional methods. Notably, they have a simple setup since they do not require vacuum equipment, can be applied to heat and vacuum-sensitive surfaces, and their effluents can be easily used on various substrates. The efficacy of APPJs arises from their non-equilibrium discharge characteristics, where radio frequency (RF) excitation predominantly heats the electrons, while ions and neutrals remain relatively cool.

Plasma jets often utilize argon or helium as carrier gas for stable plasma ignition, and are capable of producing reactive species through electron impact dissociation [11] when gases such as nitrogen [12, 13] and oxygen [14–16] are added. However, the downstream reaction dynamics in these jets are complex, involving mechanisms like excitation, ionization, chemical reactions, dissociation, and quenching. Of particular interest are reactive oxygen and nitrogen species, which are crucial for biomedical applications such as cancer [17, 18] and wound treatment [19, 20]. Effective treatment relies on understanding the optimal dosage of reactive species, necessitating precise measurement of their densities and comprehension of their production mechanisms.

Among the ROS, the $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$ states of molecular oxygen are of particular interest due to their high reactivity and, in the case of $O_2(a^1\Delta_g)$, relatively long lifetime [21]. Ozone on the other hand is a crucial precursor, not only due to its extended lifetime in gas and liquid phases but also for its ability to further react into secondary ROS, including singlet oxygen and atomic oxygen [22]. The generation and dynamics of these species in the plasma and effluent environment are therefore of high biomedical relevance.

Given the complexity of these interactions, modelling serves as a valuable tool for evaluating species interactions and densities, especially since experimental determination of several species' densities can be challenging. Zero-dimensional (0D) models provide a balance of accuracy and computational efficiency for simple gas mixtures like He/N₂ or He/O₂ [23–37]. These models can be used to study the influence of varying temperatures, gas mixture ratios, and plasma power on the plasma properties and reactive species production. For plasma jets, extending models to plug flow simulations [25, 27, 31–33] allows for spatial resolution of reactive species

along the gas flow [38], thereby describing time-dependent phenomena.

If the effect of the discharge geometry becomes significant, for example, due to a non-trivial gas flow velocity distribution, or if details of the spatiotemporal plasma dynamics are required, more sophisticated simulation methods, such as fluid [39–41] or hybrid simulations [42, 43] are employed.

Extensive studies have been conducted on various species, including reactive oxygen species [29, 42, 44] as well as hydroxyl radicals [25]. Despite these efforts, a wide range of plasma properties still need to be benchmarked against experimental data to ensure the models' predictive capability. Due to limited diagnostic access to the micro-sized discharge regions of plasma jets, measuring many species remains challenging.

The COST plasma jet [45] offers a well-defined plasma source, making it an ideal candidate for simulation comparisons. Although it has been benchmarked against various simulations with good agreement to existing data [42, 43, 46], many species' densities are yet to be validated. Further validation is essential to enhance understanding of the underlying reaction mechanisms, ultimately improving the effectiveness and reliability of atmospheric pressure plasma applications.

This paper aims to advance the understanding of the reaction kinetics of oxygen species in the COST plasma jet by providing experimental data for the difficult-to-detect species $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$, the first and second electronically excited states of molecular oxygen. Because they are heavily quenched by ozone, knowledge of its density is critical in calculating the densities of the excited molecular oxygen species. While $O_2(b^1\Sigma_g^+)$ can be measured relatively easily by emission spectroscopy in the plasma region [47], but not in the effluent region, the determination of the $O_2(a^1\Delta_g)$ density is more complicated. As a metastable state, $O_2(a^1\Delta_g)$ has a long lifetime with up to 75 min [48], due to the forbidden optical transition of the singlet state. Its emission intensity at 1270 nm is therefore low. Additionally, the thermal background in the plasma region makes it almost impossible to measure the emission signal. Therefore, mostly effluent measurements are feasible [21, 49]. To enhance the signal in the effluent, gas flow cells are used to increase the observable volume [21, 49]. Therefore, this approach is used in our study.

Here, the main focus lies on measuring the absolute densities of the excited molecular oxygen species $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$. The results were compared to two simulations, a pseudo-1D plug-flow simulation to benchmark the measured values as well as to provide insight into reaction dynamics by analysis of the simulation results, and a 2D fluid

simulation to investigate the influence of the gas flow on the reaction dynamics. The distribution of the reactive species in the gas flow cell might have a significant influence on the measured densities. The pseudo-one-dimensional (pseudo-1D) plug flow simulation, however, does not account for the spatial dynamics of the species inside the flow cell, which is inherently two-dimensional. For this purpose, the 2D fluid simulation was employed, which not only provides spatio-temporally resolved species densities, but also offers insights into the flow velocity and gas temperature. However, this simulation method is computationally more expensive, which makes comparison of broad parameter variations time-consuming. For many applications, a rough estimate of the generated densities might be enough. For these cases, the simple pseudo-1D plug flow simulation would suffice. This paper aims to use the combined insights of these methods to get a better understanding of the dynamics between ozone and the reactive species $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$. Furthermore, it tries to benchmark the oxygen species' densities and their interaction dynamics for the two commonly employed simulation models of the COST plasma jet, especially in the effluent region. Therefore, it gives insight into the optimal use cases of the two simulations and their limitations.

2. Methods

2.1. COST plasma jet

The COST micro-scaled atmospheric pressure plasma jet, which is described in detail elsewhere [5], is well characterized and capable of generating a high density of reactive species. Its plasma chemistry is described by various validated simulations, allowing for reliable comparison with experimental values. It operates at a RF of 13.56 MHz using a plane electrode configuration. The discharge channel between the electrodes is cubic. It has an area of $1\text{ mm} \times 1\text{ mm}$ and a length of 30 mm. Two quartz glass planes are glued to the sides of the electrodes and provide optical access. Inside the jet, various reactive species can be produced and transported out of the active discharge volume via the jet's nozzle. This gas stream is usually referred to as the effluent, and it can be used to treat surfaces with the produced chemical species.

2.2. Gas flow cell

The densities of many reactive species can be obtained by emission and absorption spectroscopy. These techniques can be challenging to apply in the effluent of a plasma jet because of the typically low optical emission for cross-field configuration APPJ's [50], small observable volume [21, 49] and short absorption length for the effluent dimensions [51]. To achieve a good signal-to-noise ratio for spectroscopic measurements, a large detection volume or long optical paths are favourable. This is especially critical for low emissivity from long-lived species like $O_2(a^1\Delta_g)$ [21, 49].

To this end, a gas flow cell was used to contain the effluent and increase the detection volume, in which light could

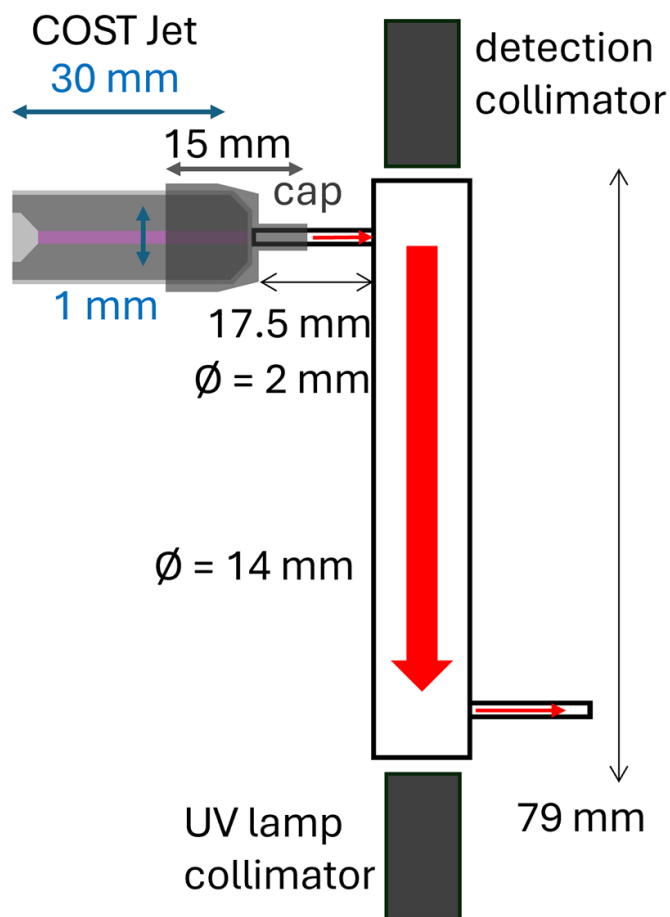


Figure 1. Schematic of the quartz flow cell dimensions and the connection to the COST jet, as well as the position of the collimators for the optical setups. The red arrows indicate the gas flow direction.

be emitted or absorbed, respectively. The use of a gas flow cell does not allow for an analysis of the spatial distribution of species in the effluent region. However, it provides an estimate of the densities produced inside the plasma jet if the species is long-lived. The cell's schematic is shown in figure 1. It is made out of quartz glass to allow the transmission of UV-light for absorption spectroscopy. The gas flow cell is connected to the COST plasma jet by a plastic cap, which guides the effluent from the cubic discharge channel of the jet into a 17.5 mm long round tube with a diameter of 2 mm. The effluent then reaches the cylindrical gas flow cell with an inner diameter of 14 mm and a length of 79 mm. The plane windows at the ends of the flow cell allowed optical access. Attached to the outlet of the gas flow cell was a long pipe to prevent the backflow of air into the cell.

2.3. Absorption spectroscopy of ozone

The optical setup of the absorption spectroscopy measurement is shown in figure 1. The utilized light source for the absorption spectroscopy was a laser-driven broad band lamp (Energetiq EQ-99 LDLS). The light was guided through a

fibre attached to the UV lamp and a collimator, which parallelized the light to a beam diameter of 13 mm. This matched the gas flow cell dimensions. The light passed through a narrow bandpass UV filter (Edmund optics bandpass filter, OD 4, CW = 254 nm, FWHM = 10 nm). This wavelength range fits the absorption of the Hartley band of ozone while reducing the energy input of the UV light, which could lead to dissociation of O_2 and thus further ozone formation. The intensity of the absorption spectra was calculated from the averaged intensity within the filter wavelength range (approx. 250–260 nm). An example ozone spectrum is shown in the appendix figure 16. At the end of the gas flow cell, another collimator and optical fibre were used to guide the light into a UV sensitive spectrometer (Avantes AVASPEC-ULS-2048x64TEC-EVO, UB from 200 nm to 720 nm; resolution 0.3 nm).

To measure the density of $O_2(a^1\Delta_g)$, knowledge of the ozone density is crucial due to its significant quenching effect on $O_2(a^1\Delta_g)$. The ozone density was determined using absorption spectroscopy, with all spectra having an integration time of 1 s. It was ensured beforehand that the signal remained stable over time by monitoring it for 1 h, confirming consistency once the jet was heated up. An initial spectrum of the broadband lamp was recorded just before igniting the plasma (I_0), followed by spectra taken every minute for 5 min after plasma ignition (I_T), ensuring the measurement was performed with the discharge in an equilibrium state. After each measurement, a minimum of 5 min was allowed for the absorption signal to reset to an undetectable level. The ozone density was calculated using Lambert–Beer’s law using the following formula [52] and averaged over the 5 spectra within the measurement time of 5 min,

$$n_{O_3} = \frac{p}{k_B \cdot T} \cdot \frac{\log\left(\frac{I_0}{I_T}\right)}{k \cdot d} \quad (1)$$

where p is the ambient pressure, T the effluent temperature in the jet (approx. 300 K), I_0 the initial intensity of the broad band lamp passing through the filter, I_T the transmitted intensity through the gas flow cell, k the absorption coefficient of ozone ($k = 333 \text{ cm}^{-1}$) [53] and d the length of the gas flow cell ($d = 79 \text{ mm}$).

2.4. Emission spectroscopy of $O_2(a^1\Delta_g)$

Figure 1 shows the optical setup for the $O_2(a^1\Delta_g)$ detection. The emission of the $O_2(a^1\Delta_g)$ at 1270 nm was collected by the detection collimator and guided through an optical fibre into an infrared sensitive spectrometer (Avantes AVASPEC-NIR512-1.7HSC-EVO, NIR150-1.2 from 900 nm to 1695 nm; wavelength resolution 0.3 nm). The $O_2(a^1\Delta_g)$ spectrum is shown in the appendix figure 17. The collimator had a diameter of approx. 10 mm, which allowed to gather the emission passing through the plane quartz windows of the gas flow cell. To calculate the $O_2(a^1\Delta_g)$ density the spectrometer had to be calibrated beforehand. To this end, a laser diode (RLT1270-20MGS-B; PW = 1270 nm, FWHM = 0.3 nm) was used, for which the intensity was measured and compared to the radiative power given by an

Table 1. Quenching coefficients of different reactive species with $O_2(a^1\Delta_g)$ calculated using their respective densities and rate coefficients. Rate coefficients are taken from [38].

Reaction	q_i / s^{-1}	$k_i / \text{cm}^3 \text{s}^{-1}$	n_i / cm^{-3}
$O_2(a^1\Delta_g) + O_2 \rightarrow 2 O_2$	0.1	1.5×10^{-18}	5.0×10^{16}
$O_2(a^1\Delta_g) + O \rightarrow O_2 + O$	0.8	2.0×10^{-16}	4.0×10^{15}
$O_2(a^1\Delta_g) + O_3 \rightarrow O + 2 O_2$	12.1	4.0×10^{-15}	3.0×10^{15}
$O_2(a^1\Delta_g) + He \rightarrow O_2 + He$	0.1	1.0×10^{-21}	1.0×10^{19}

absolutely calibrated InGaAs-photodiode (Thorlabs FGA21-CAL; wavelength range: 800–1700 nm). First, the intensity was measured using the spectrometer. Keeping the optical setup the same, the fibre was then coupled into the calibrated photodiode. This allowed us to calculate the energy-intensity calibration constant $C = \frac{I_0}{P \cdot t_{\text{int}}}$, where I_0 is the peak intensity of the laser diode at 1270 nm, P the measured power of the calibrated photodiode and t_{int} the integration time of the spectrometer.

To calculate the $O_2(a^1\Delta_g)$ density, the following equation [21, 47, 54] was used,

$$n_{O_2(a^1\Delta_g)} = I \cdot C \cdot \frac{1}{A_{ik} \cdot Q} \cdot \frac{\lambda}{h \cdot c} \cdot \frac{1}{V} \cdot \frac{1}{f(d\Omega) \cdot g} \quad (2)$$

where I is the intensity, C is the wavelength dependent energy-intensity calibration constant of the spectrometer, V the volume of the flow cell. $f(d\Omega) = 2.7 \times 10^{-3}$ the geometric factor of how many photons produced inside the gas flow cell are reaching the detector and $g = 0.63$ the loss factor of the optical components (estimated through the transmission of the optical components). A_{ik} is the Einstein coefficient of $O_2(a^1\Delta_g)$ ($A_{ik} = 2.22 \times 10^{-4} \text{ s}^{-1}$ [48]). $Q = \frac{A_{ik}}{A_{ik} + q}$ is the photon yield, which relates the observable radiative transition to the non-radiative decay of the $O_2(a^1\Delta_g)$ molecule. $q = \sum_i k_i n_i$ is the quenching rate.

Table 1 shows the quenching rates of the most prominent quenching partners for $O_2(a^1\Delta_g)$. For our case, ozone possesses by far the highest rate coefficient, which simplifies the expression to $q = k_{O_3} \cdot n_{O_3}$, with k_{O_3} being the quenching rate coefficient of ozone with $O_2(a^1\Delta_g)$ and n_{O_3} the ozone density. The previously acquired data for the ozone density of the experiment is used to calculate the absolute $O_2(a^1\Delta_g)$ density.

2.5. $O_2(b^1\Sigma_g^+)$ density measurement by emission spectroscopy

The setup for the $O_2(b^1\Sigma_g^+)$ emission spectroscopy is shown in figure 2. The flow cell setup prevented optical access to the last third of the jet’s plasma chamber due to the cap. Therefore, the $O_2(b^1\Sigma_g^+)$ density measurement was conducted without the flow cell. As $O_2(b^1\Sigma_g^+)$ is quenched quickly outside of the plasma through ozone, its density is measured almost exclusively inside the plasma chamber. This change in setup should have only a minimal influence. $O_2(b^1\Sigma_g^+)$ emits at 760 nm and was captured with the infrared sensitive

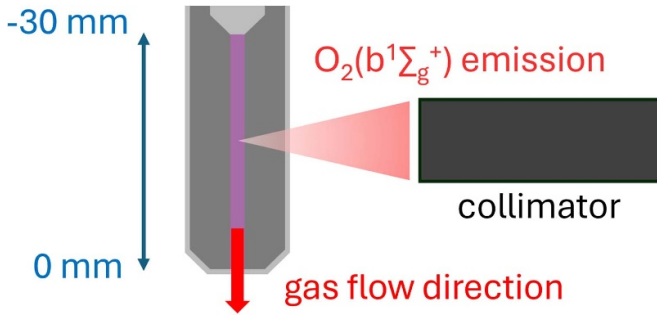


Figure 2. Emission spectroscopy setup for the $O_2(b^1\Sigma_g^+)$ measurement in the jet. Red arrow indicates gas flow direction.

spectrometer used in the $O_2(a^1\Delta_g)$ measurement ($O_2(b^1\Sigma_g^+)$ spectra shown in appendix figure 18). The emission was measured mainly in the discharge channel of the COST plasma jet. Quick quenching of $O_2(b^1\Sigma_g^+)$ quickly reduces its density beyond the detection threshold outside of the plasma jet.

To measure the $O_2(b^1\Sigma_g^+)$ density, the spectrometer had to be energy-intensity calibrated similarly as described in the $O_2(a^1\Delta_g)$ section ($A_{ik} = 0.08 \text{ s}^{-1}$). Here, an LED (Thorlabs LED760L; $PW = 760 \text{ nm}$, $FWHM = 24 \text{ nm}$) was employed instead of a laser diode. An optical filter ($CW = 760 \text{ nm}$, $FWHM = 10 \text{ nm}$) was placed between diode and spectrometer, to get a more comparable power measurement between calibrated diode and spectrometer, as the width of the $O_2(b^1\Sigma_g^+)$ spectrum is similar to the spectral width of the filter. The intensity of the spectrometer signal was then integrated over the spectral width and compared to the energy of a calibrated Si-photodiode (Thorlabs FDS100; wavelength range: 350–1100 nm). The optical setup was unchanged when switching the fibre between spectrometer for the intensity measurement and the photodiode for the power measurement.

3. Computational methods

The two simulations presented use a very similar chemical kinetics scheme, leading to a similar prediction about the species distribution inside the plasma chamber. Only simplified wall reactions were taken into account for the reactive species or, in the case of ozone, were completely neglected. The used wall reaction rates were optimized for the plasma region of the COST jet. Investigation of the correct wall rate coefficient for various species was beyond the scope of this study.

3.1. Pseudo-1D plug flow model

Pseudo-1D plug flow model is used in this study to calculate the spatially resolved species' densities along the gas flow direction in the jet chamber [46, 55]. The particle balance equations and electron energy balance equation of the zero-dimensional simulation are solved to output the time-resolved species densities and an effective electron temperature in an

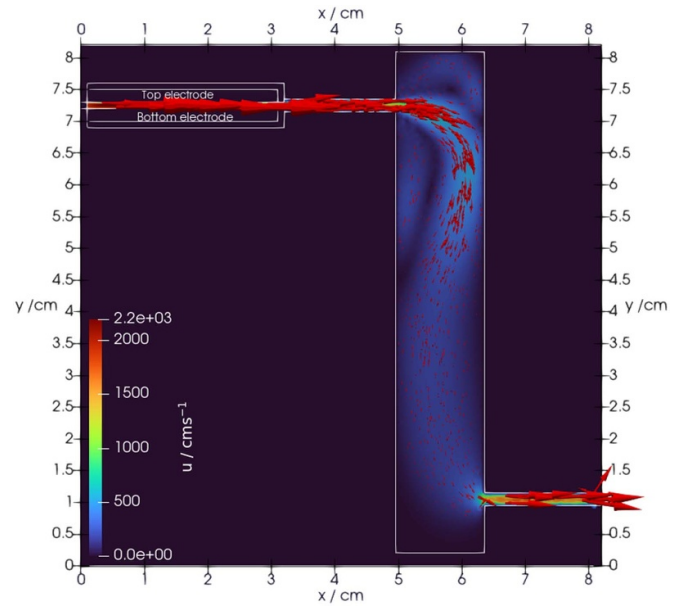


Figure 3. Computational domain and spatial distribution for the gas velocity at 1 slm He flow. The colour bar shows the gas velocity while the red arrows indicate the gas flow direction.

infinitesimal plug volume. This plug volume co-moves with the gas flow, which allows the temporal variation to be converted into a spatial variation through knowledge of the gas flow velocity.

The gas temperature in the jet chamber is varied as a function of the absorbed power in the simulations, using equation (1) in [55]. The gas temperature of the feed input gas and that in both effluent regions (i.e. 17.5 mm long tube and 79 mm long gas flow cell in figure 1) are approximated to be 300 K. The pressure is assumed to be $1 \times 10^5 \text{ Pa}$ in all regions.

The plug flow model calculations in both effluent regions are carried out in a similar way to those in the jet chamber. However, the model is modified due to the different geometry and power between the chamber and effluent. In particular, the cross section area of the rectangular jet chamber ($1 \times 1 \text{ mm}^2$) is smaller than those of both cylindrical effluent regions ($1 \times 1 \pi \text{ mm}^2$ and $7 \times 7 \pi \text{ mm}^2$). Because of this, the flow velocity in both effluent regions is calculated by replacing the gas temperature, pressure and cross section area of the jet chamber in equation (5) in [46], with those of the corresponding effluent region in the current study. Therefore, the gas flow velocity in the jet chamber is larger than those in both effluent regions, e.g. at 1 slm He diluted with 0.5% O_2 , the flow velocities in the jet chamber, the 17.5 mm long entry tube and 79 mm long gas flow cell are calculated to be around 1882 cm s^{-1} , 533 cm s^{-1} and 11 cm s^{-1} , respectively. The flow velocity value used in plug flow model in the jet chamber is similar to that calculated by the fluid model presented in figure 3, while the velocities of the plug flow model in both effluent regions underestimate those calculated by the fluid model. The underestimation in the 79 mm long gas flow cell is likely ascribed to the fact that the gas flow direction in the gas

flow cell region assumed in the plug flow model is the same as those in the jet chamber and tube (i.e. it is not perpendicular to those in the jet chamber and tube, as illustrated by the experimental setup shown in figure 1). This is less realistic compared to the gas flow direction simulated by the fluid model.

The reaction rate of a neutral species at the wall is calculated by replacing the effective diffusion length [56], the volume and the net surface area with those for the corresponding effluent region of the gas flow cell setup. Furthermore, the electron energy balance equation (i.e. equation (9) in [46]) is excluded in the simulations of both effluent regions due to the absence of the power and the resulting negligible effect of the electron density. Therefore, only the particle balance equations are solved in the simulations of both effluent regions.

A Boltzmann solver based on the steady-state solution of the Boltzmann equation under the two-term approximation, i.e. the open-source simulation tool Lisbon Kinetics Boltzmann published by Tejedo-del-Caz *et al* [57], is used to calculate the non-Maxwellian electron energy distribution functions (EEDFs) for a range of values of the applied reduced electric field.

The plasma-chemical kinetics scheme considered in this work is identical to that used in [55], where the lists of all the species and the reactions are provided. Specifically, He, He(2^3S), He $_2^*$, He $^+$, He $_2^+$, O(3P), O $_2$ ($v=0$), O $_3$, O(1D), O $_2$ ($a^1\Delta_g$), O $^+$, O $_2^+$, O $_4^+$, O $^-$, O $_2^-$, O $_3^-$, O $_4^-$, e $^-$, and O $_2$ ($b^1\Sigma_g^+$) are included. It is worth to note that the simulated O(3P) densities produced in He/O $_2$ plasma jets in [55] are well validated against measured densities from multiple publications under variations of the absorbed power, gas flow rate, and mixture ratio.

3.2. Fluid model

The computational model employed in this study to obtain spatially resolved data in two dimensions is *nonPDPSIM*, a fluid-based plasma simulation code [58, 59]. The code solves the continuity equation for charged and neutral species, assuming the drift-diffusion approximation and the compressible Navier–Stokes equations, providing the species densities, flow field and the gas temperature distribution, on an unstructured triangular grid. The charged species considered in the model are electrons (e $^-$), positive (O $_2^+$, O $^+$, He $^+$), and negative ions (O $^-$, O $_2^-$, O $_3^-$), while neutral species include He, O $_2$, O, O $_3$, O $_2$ ($v=1-4$) (first four vibrational levels of O $_2$), O $_3$ (v), O $_2$ ($a^1\Delta_g$), O $_2$ ($b^1\Sigma_g^+$), O(1D), and He * (which is an ensemble of He(2^3S) and He(2^1S)). The transport coefficients for heavy species (i.e. not electrons) are calculated by assuming that the respective species temperature is equal to the gas temperature. The electron transport coefficients (mobility and diffusion) are calculated from a 2-term Boltzmann solver. The simulation uses the local mean energy approximation and therefore solves the energy equation as well. The chemistry set and the cross sections for the Boltzmann solver are identical to those reported in [60]. Note that the reaction set used here differs slightly

from that of the plug flow model. However, as both have been independently validated under COST jet conditions, and since the primary goal of this work is to compare simulation results with experimental data, we consider the use of different yet well-established chemistry sets to be acceptable.

The spatial resolution of the unstructured mesh was adapted to the relevant physical scales of each region. Inside the jet, the grid spacing was $3 \cdot 10^{-5}$ m in the y -direction (between the electrodes) and $7 \cdot 10^{-4}$ m along the x -axis, as the variation is expected to be stronger across the gap. In the connecting tube leading from the jet to the flow cell, the resolution was $2 \cdot 10^{-4}$ m, while in the flow cell, it was again coarsened to $7 \cdot 10^{-4}$ m, reflecting the reduced gradients and weaker dynamics in this region.

Figure 3 illustrates the computational domain along with the spatial distribution of the gas flow field at a He inflow of 1 slm after the stationary state has already been attained. The neutral species generated within the jet (with the ‘Top electrode’ driven and the ‘Bottom electrode’ grounded) flow through a tube with a 2 mm gap length, before entering the flow cell, as shown in the figure. The dimensions of the computational domain agree with the experimental setup described above. As seen in the figure, the gas velocity distribution is non-uniform in the flow cell. Note that, due to the change in gap length between the jet electrodes and the tube leading to the flow cell, the velocity decreases by a factor of 2 (as the simulation is two-dimensional). Moreover, as the gas enters the flow cell, its velocity further decreases, forming two vortices near the inlet.

All simulations were run for a total physical time of 0.2 s for the neutral species and flow field, while the plasma dynamics were resolved over 30 μ s using a time-slicing approach [59]. This corresponds to roughly 10 flow-through times of the domain, which is sufficient for convective transport. However, full diffusive homogenization, particularly for long-lived species such as ozone, would require significantly longer runtimes (i.e. multiple seconds) due to slower mixing. Although most species, including ions and short-lived neutrals, approach convergence within the simulated time, long-lived species such as ozone, and to a lesser extent O $_2$ ($a^1\Delta_g$), which is affected by its interaction with ozone, continue to evolve slightly due to incomplete diffusive mixing. Based on the observed convergence behaviour, we estimate that the deviation in their volume-averaged densities at the end of the simulation is on the order of 10%–15%. The current simulations, while not fully converged in the flow cell, nonetheless capture the key trends and magnitudes relevant for comparison with experiment.

4. Results

4.1. Ozone density

The ozone density was studied under varying power, flow rate, and O $_2$ admixture conditions. Experimental results were compared with simulations from both the pseudo-1D plug flow model and the 2D fluid model. The plug flow model focuses

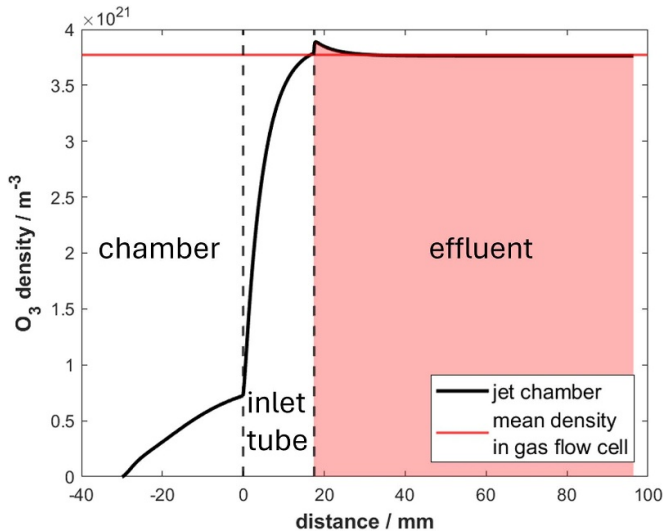


Figure 4. O_3 density calculated by the plug-flow model along the gas flow direction in and outside the plasma jet. Averaged density in the gas flow cell volume is shown as a red line. Dashed lines show the transition from the jet (left) to the inlet tube (middle) and to the flow cell (right). An effluent temperature of 300 K, a plasma power of 1 W, a 1 slm He flow, and a 0.5 % oxygen admixture are assumed.

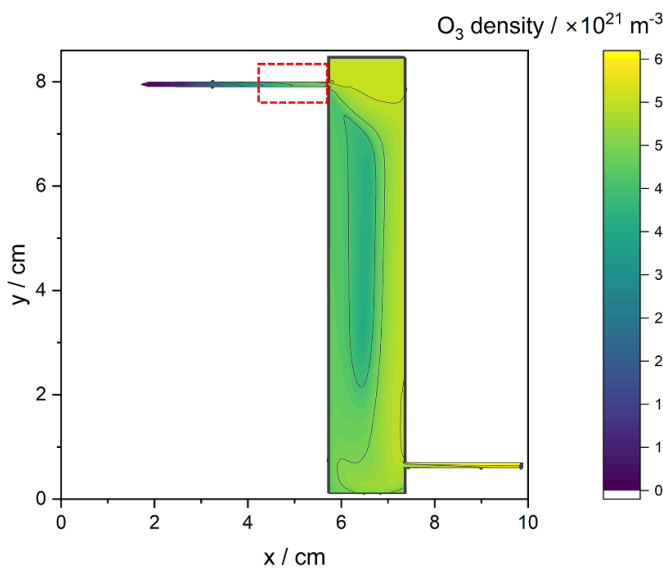


Figure 5. O_3 density distribution inside the gas flow cell simulated by a fluid model for an effluent temperature of 300 K, plasma power of 1 W, flow of 1 slm He, and a 0.5 % oxygen admixture. The red dotted area marks the inlet tube after the plasma jet channel.

on plasma chemistry within the jet and effluent, as illustrated in figure 4, while the 2D fluid model accounts for both chemistry and gas dynamics in the flow cell geometry, as shown in figure 5.

Figure 4 shows the ozone density calculated by the plug flow model along the gas flow direction, where $x = 0$ m is the end of the jet electrodes. In the plasma channel, the ozone density increases asymptotically, reaching a maximum at the jet nozzle. Along the length of the jet, oxygen is dissociated into atomic oxygen. The atomic oxygen then reacts with

molecular oxygen to form ozone, which is one of the most prevalent production mechanisms. With the increased dissociation degree along the jet, more atomic oxygen is available to produce ozone [61]. The production would eventually equalize with the reaction of ozone with $\text{O}_2(\text{b}^1\Sigma_g^+)$. However, for a flow of 1 slm, the residence time in the plasma region is not long enough to reach an equilibrium.

In the inlet tube to the gas cell, the ozone density continues to rise asymptotically. The fast rise, with respect to distance, of the ozone density from the transition of the jet chamber to the inlet tube occurs because of the slower gas velocity in the inlet tube. This leads to a strong increase in ozone density with distance as the residence time in the inlet tube is longer for the same path length. Furthermore, ozone destruction rates, like reactions with $\text{O}_2(\text{b}^1\Sigma_g^+)$ or electron impact dissociation, are lower in the effluent region.

Upon entering the flow cell, the gas velocity decreases further, which leads to a spike in the density because of the significantly higher residence time. Due to the minimal influence of destruction reactions and ozone's long lifetime, the density remains approximately constant. The plasma chemistry, therefore converges to the more long lived species like ozone and molecular oxygen. The ozone density in the flow cell is averaged along the flow direction vector, as shown in figure 4. This averaged value is then compared to the experimental results.

Figure 5 shows the two dimensional ozone density distribution in the gas flow cell as simulated by the 2D fluid model. The effluent is then directed into a tube that connects vertically to the gas flow cell, as described in section 2.2. As shown in the plug flow simulation, the ozone density increases significantly outside the plasma zone, especially in the flow cell. In the cell, the flow follows a vortex along the cell walls, resulting in a maximum density at the outer sides of the flow cell. In the centre axis of the cell, the density is lower due to the vortex effect. Additionally, the pocket at the entrance, where the optical window is located, also generates a vortex, leading to a slightly higher density in that area compared to the rest of the cell. The inhomogeneous distribution of ozone by the vortex is a result of the relatively short simulation time (≈ 0.2 s). With a prolonged simulation time, the modelled density of long-lived oxygen species like ozone is expected to reach a more homogeneous distribution inside the cell. Based on the convergence behaviour observed in our simulations, we estimate that the remaining deviation in the volume-averaged ozone density at the end of the 0.2 s simulation is approximately 10%–15%. A similar magnitude of deviation is expected for $\text{O}_2(\text{a}^1\Delta_g)$, due to its chemical coupling with ozone. This is primarily due to diffusion: since the diffusion coefficient of ozone in helium is $D_{\text{O}_3} = 7.13 \cdot 10^{-5} \text{ m}^2 \text{ s}^{-1}$ [11], the timescale for homogenization would be on the order of seconds. Unfortunately, increasing the simulation time to a sufficient amount was computationally very expensive and beyond the scope of this study.

Figure 6 shows the ozone density as a function of plasma power for a helium flow of 1 slm and an oxygen admixture of 0.5%. Both simulations and experimental results demonstrate a gradual increase in ozone density with rising plasma power. The increased plasma power leads to a higher dissociation degree of molecular oxygen and therefore to a higher ozone

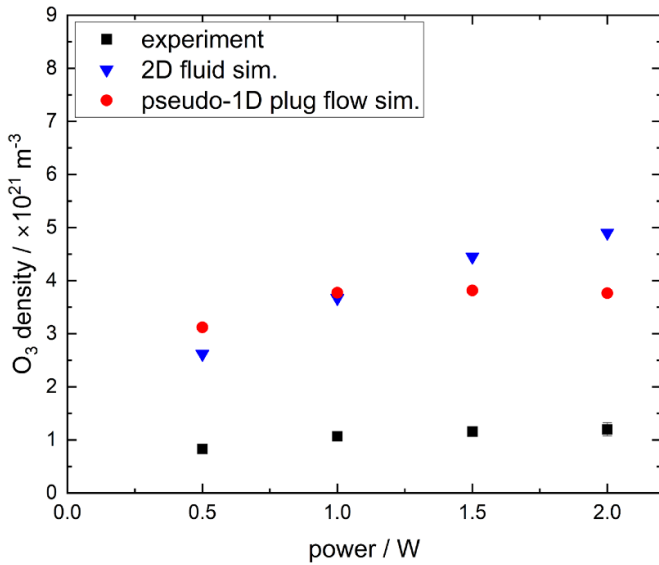


Figure 6. O₃ density comparison between simulation and experiment for an effluent temperature of 300 K, flow of 1 slm He, and a 0.5% oxygen admixture, depending on the plasma power.

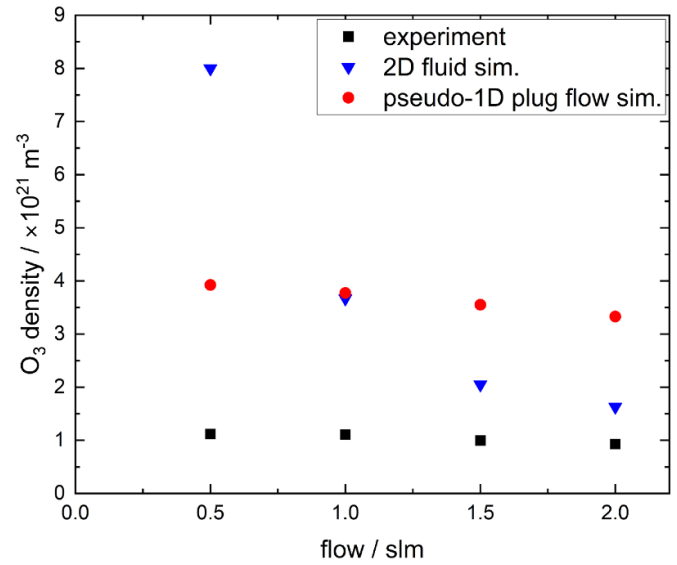


Figure 7. O₃ density comparison between simulation and experiment for a effluent temperature of 300 K, 1 W plasma power and a 0.5% oxygen admixture, depending on the helium flow.

density as more atomic oxygen is available. The experimental data show a linear increase of the ozone density with power. This trend is matched by the 2D fluid simulation, although its slope is higher than that observed for the experimental values. The density values of the two simulations match quite well, but are approx. a factor of three higher than what the experimental data suggests, though they remain within the same order of magnitude. The plug flow simulation also shows a more asymptotic increase. The ozone density reaches a plateau at 1 W. Here, the ozone production is likely equilibrated by reactions with $\text{O}_2(b^1\Sigma_g^+)$, which prevents the ozone density from increasing further. The good agreement in the density values of the two simulations supports the assumption that the volume-averaged density in the plug flow simulation gives a reasonable agreement under the condition that the density is homogeneously distributed within the gas flow cell. One possible explanation for the discrepancy between the simulations and experimental results could be related to the reaction dynamics within the flow cell. For instance, the simulations do not account for ozone's wall destruction reactions. Given the similarity in the simulated ozone densities, it is plausible that both simulations underestimate the destruction rate of ozone and/or overestimate its production, resulting in higher simulated values compared to the experiment. Additionally, ozone is sensitive to conditions such as temperature. Small deviations in experimental temperature from the assumed 300 K effluent temperature in the simulations could also contribute to the lower ozone density observed experimentally [62, 63].

The ozone density is influenced by the gas flow velocity, as shown in figure 7. Both experimental results and simulations agree that the ozone density is highest at low flow rates and decreases as gas velocity increases. This is due to the longer residence time in the plasma volume at lower flows. Ozone is primarily formed through the reaction of atomic oxygen with molecular oxygen, in the presence of a third body. With a

lower gas flow, the longer residence time in the plasma region leads to a higher dissociation degree of oxygen, increasing the atomic oxygen density and, consequently, the ozone density. Both the experiment as well as the plug flow simulation show a linear decrease with the flow velocity with an almost identical slope. The 2D fluid simulation, however predicts a more asymptotic decrease. An explanation might be that the fluid simulation does not reach a homogeneous distribution in the gas flow cell within the simulation time, as shown in figure 5. This explains the high discrepancy for low flows, as here the time to reach an equilibrium is much higher than for higher flows. Higher flows, on the other hand, should reach the equilibrium faster and indeed have a smaller deviation from experimental data. As observed previously in the power variation measurements, the simulations still overestimate the ozone density compared to the experiment. It should, however, be noted that the 2D fluid simulation density values come very close to the experimental values for high flows, as the ozone distribution gets more homogeneous. With longer computation time, the 2D fluid simulation might therefore be able to provide an accurate description of the experimentally measured densities.

Figure 8 shows the ozone density as a function of oxygen admixture. Both simulation and experimental results demonstrate an ozone density increase with the oxygen admixture. Ozone is a product of the recombination of atomic oxygen with molecular oxygen. It is well known that the COST jet has an optimal oxygen admixture for atomic oxygen production around 0.5 % in helium [61]. Lower admixtures result in a lower atomic oxygen density because less molecular oxygen can be dissociated, while higher admixtures reduce the atomic oxygen density because the dissociation of molecular oxygen is less efficient and recombination to ozone occurs more rapidly. Even when the atomic oxygen density decreases with higher oxygen admixtures, the ozone density still rises

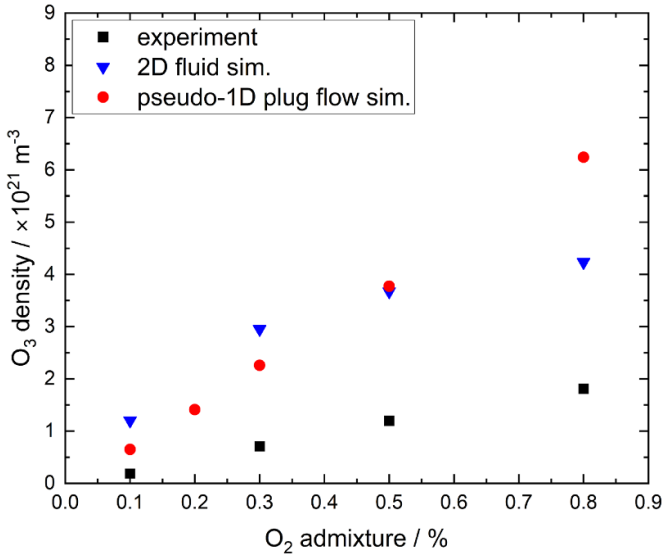


Figure 8. O₃ density comparison between simulation and experiment for a effluent temperature of 300 K, 1 W plasma power and a helium flow of 1 slm, depending on the oxygen admixture.

slightly for higher admixtures, as atomic oxygen has more molecular oxygen it can react with. Both simulations predict a rise in the ozone density, which agrees with the experimental data. Furthermore the ozone density values predicted by the two simulations fit quite well with the experimental results for low admixtures. High admixtures, however, lead to an overestimation of the ozone density. The 2D fluid simulation provides a more accurate description of the dependency on the admixture observed in the experimental results, with a more asymptotic increase. The plug flow simulation, however, predicts an almost linear increase in the ozone density, which, especially for high flows, leads to a discrepancy to the experimental values. This indicates that the species distribution induced by the gas dynamics may play a more important role for high oxygen admixtures as low admixtures generally have a better agreement with the experimental values.

Both simulations provide a good estimate of the measured densities within the same order of magnitude, although overestimating the ozone density by a factor of around three. Still, the 2D fluid simulation is able to reproduce the more complex dependencies of the ozone density on the admixture variation and, given more time to converge, maybe also for the flow variation, as higher flows have shown a better agreement with experimental results. The plug flow simulation was able to replicate the trends of the power variation and flow variations, but represents the trend in the admixture variation less accurately.

4.2. O₂(a¹Δ_g) density

The experimentally determined O₂(a¹Δ_g) density was compared to the plug flow simulation and the 2D fluid simulation as with the ozone density.

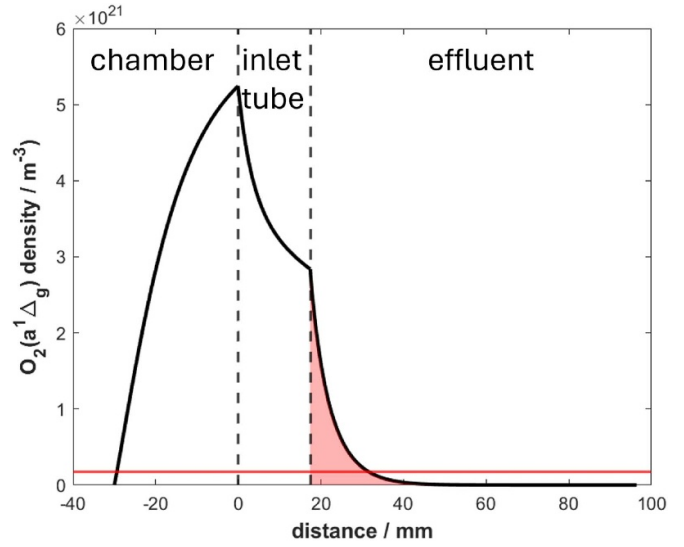


Figure 9. O₂(a¹Δ_g) density calculated by the plug flow model along the jet and effluent in the gas flow cell. Dashed lines show the transition from the jet (left) to the inlet tube (middle) and to the flow cell (right). Averaged density for the gas flow cell volume is shown as a red line. An effluent temperature of 300 K, plasma power of 1 W, 1 slm He flow, and a 0.5% oxygen admixture are assumed.

The simulated O₂(a¹Δ_g) density for the plug flow model is shown in figure 9. It can be observed that within the discharge, the O₂(a¹Δ_g) density rises asymptotically towards the end of the jet. From there the gas flow transitions into the wider inlet tube ($d = 2$ mm) to the flow cell as indicated by the left dashed line. The flow cell begins after the second dashed line. In the effluent, the O₂(a¹Δ_g) decays exponentially as it reacts. This behaviour can be observed for multiple species, like atomic oxygen [61]. The decay rate, with respect to distance, increases when entering the gas flow cell. The bigger cross section in the flow cell results in a slower gas velocity, so the O₂(a¹Δ_g) decays faster over the same length scale.

To compare the plug flow model density to the experimental values, the O₂(a¹Δ_g) density was averaged along the gas flow direction after the inlet tube, as shown by the red area in figure 9. The O₂(a¹Δ_g) density is not homogeneously distributed inside the flow cell, but later comparison will show that this assumption still leads to a reasonably good agreement between plug flow simulation and experimental results.

Figure 10 shows the density distribution of the O₂(a¹Δ_g) inside the gas flow cell geometry. The O₂(a¹Δ_g) decays along the entrance tube to the flow cell outside the plasma before it reaches the cell. As the lifetime of O₂(a¹Δ_g) is shorter than that of ozone, and it reacts at a high rate with ozone, the distribution of O₂(a¹Δ_g) is more inhomogeneous. Its density also generally decreases with increasing distance from the inlet. At the point of entering the flow cell, the O₂(a¹Δ_g) density is around $2.4 \times 10^{21} \text{ m}^{-3}$, which is in good agreement with the plug flow simulation values shown in figure 9 where the density is around $3.0 \times 10^{21} \text{ m}^{-3}$. At the point of leaving the flow cell, the density is almost halved to around

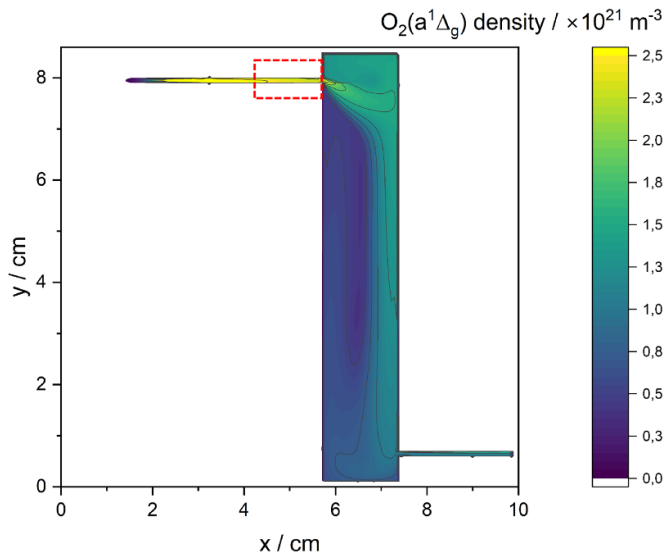


Figure 10. Two-dimensional $O_2(a^1\Delta_g)$ density in the gas flow cell volume calculated by the 2D fluid simulation of the COST plasma jet for effluent temperature of 300 K, power of 1 W, 1 slm He flow and a 0.5% oxygen admixture.

$1.3 \times 10^{21} \text{ m}^{-3}$, which is much higher than the plug flow simulation predicts because of the slow gas velocity. As observed for ozone, the $O_2(a^1\Delta_g)$ density along the flow cell wall opposite to the entrance tube is slightly higher than on the entrance side. The $O_2(a^1\Delta_g)$ therefore follows the vortex with a decrease of the density in the middle of the cell. To compare the results to the experiment, the density was averaged over the volume of the gas flow cell as described for the determination of the ozone density.

To study the $O_2(a^1\Delta_g)$ density produced by the COST plasma jet we conducted variations of the plasma power, gas flow, and oxygen admixture.

Figure 11 shows the $O_2(a^1\Delta_g)$ density depending on the plasma power from experiment and simulation. The 2D fluid simulation and the experiment show an increase in the density with power. As $O_2(a^1\Delta_g)$ is produced mostly through electron impact excitation and the reaction of $O_2(b^1\Sigma_g^+)$ with ozone, it is reasonable that the density increases with higher plasma power since this also increases the ozone and $O_2(b^1\Sigma_g^+)$ densities.

The experimental data shows a steady increase of the $O_2(a^1\Delta_g)$ density with the power. The same is observable for the 2D fluid simulation, although the values are roughly three times higher. However, the slope is matching very well. As mentioned before, the ozone distribution inside the flow cell does not reach a steady state within the short simulation time. This inhomogeneous distribution of the ozone might lead to a locally reduced reaction rate with $O_2(a^1\Delta_g)$ and may result in a higher average density.

The densities calculated from the plug flow simulation are within the same order of magnitude as those of the experiment and the 2D fluid simulation. However, they show only a small dependency on the plasma power, which mainly results from

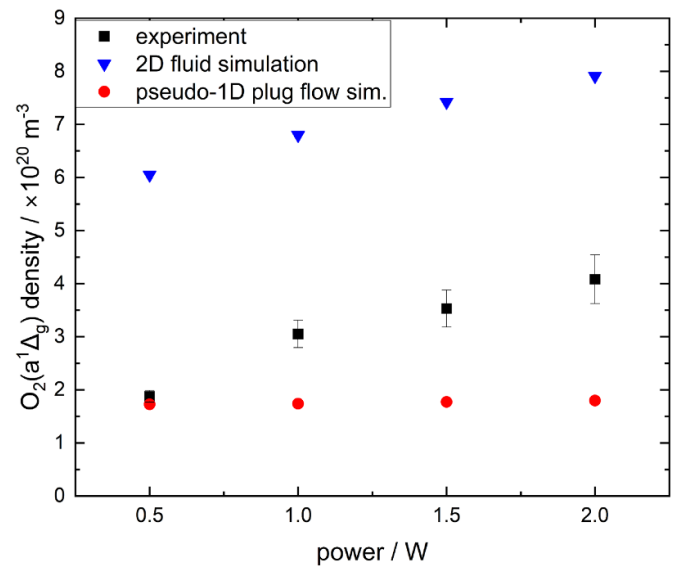


Figure 11. $O_2(a^1\Delta_g)$ density comparison between simulation and experiment for an effluent temperature of 300 K, power of 1 slm He flow and a 0.5% oxygen admixture as a function of the plasma power.

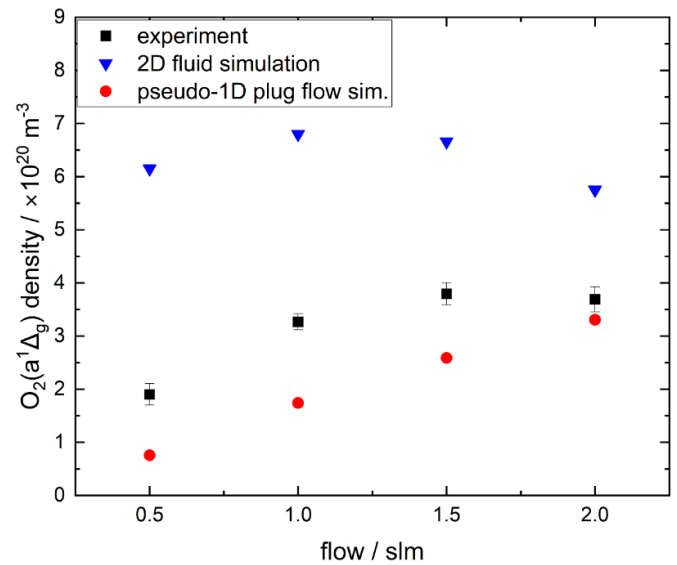


Figure 12. $O_2(a^1\Delta_g)$ density comparison between simulation and experiment for an effluent temperature of 300 K, plasma power of 1 W, and a 0.5% oxygen admixture, depending on the gas flow.

the slow gas velocity in the gas flow cell. Due to the long residence time in the cell and the inlet tube, most of the $O_2(a^1\Delta_g)$ already decayed. This leaves only the tail of the exponential distribution, which is not impacted strongly by an increase in the initial $O_2(a^1\Delta_g)$ density produced inside the plasma channel.

In figure 12 the $O_2(a^1\Delta_g)$ density is shown for a variation of the gas flow. The experimental results show an increasing $O_2(a^1\Delta_g)$ density with the gas flow, reaching a maximum at 1.5 slm and then slightly decreasing towards 2.0 slm. The shorter residence time due to the increased flow velocity gives

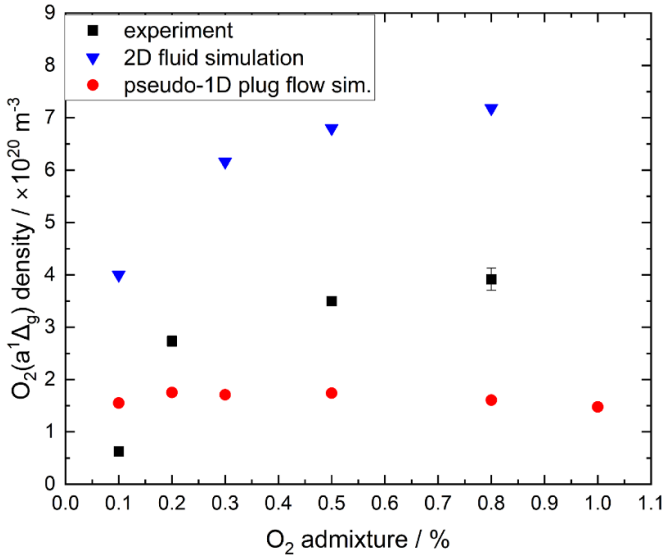


Figure 13. O₂(a¹Δ_g) density comparison between simulation and experiment for an effluent temperature of 300 K, plasma power of 1 W, and 1 slm He flow, depending on the oxygen admixture.

the O₂(a¹Δ_g) less time to be quenched, thus more O₂(a¹Δ_g) is reaching the flow cell, increasing its average density. On the other hand, with the increased gas velocity, the residence time in the plasma region decreases, which leads to less O₂(a¹Δ_g) being formed. These two effects appear to cancel out for flow rates of 1.5 slm to 2.0 slm. After that, it is reasonable to assume that the density would decrease, as less and less O₂(a¹Δ_g) can be produced in the plasma volume with the decrease in residence time.

The 2D fluid simulation shows an increase in the O₂(a¹Δ_g) density from 0.5 slm to 1.0 slm, where it stays mostly constant before declining after 1.5 slm. This trend can be explained similarly to the experimental results. However, in case of the fluid simulation an earlier decline in the density can be observed, which is most likely again due to the species distribution not reaching a complete equilibrium state within the simulation time.

The plug flow simulation shows more of a linear increase in the density, with its values being slightly lower than the experimental results. Because of the low gas flow velocity in the flow cell, an increase in the gas flow seems to have a more pronounced influence on the averaged O₂(a¹Δ_g) density than the decrease in production due to the shorter residence time in the plasma volume. This results in a more linear increase in the density. Flows beyond 2 slm might still lead to decreased densities if the residence time in the plasma volume continues to decrease.

Figure 13 shows the impact of the oxygen admixture on the O₂(a¹Δ_g) density. The plasma power was kept constant at 1 W. Increasing the oxygen percentage in the gas flow leads to an asymptotic increase of the O₂(a¹Δ_g) density in the effluent for the experimental results. This is likely due to the higher likelihood of electron impact excitation in the jet chamber if more oxygen molecules are present. The 2D fluid simulation

follows the experimental trend quite closely, but has slightly higher density values.

The plug flow simulation does not predict a strong dependence on the oxygen admixture. Only from 0.1% to 0.2% a slight increase in the density is visible, while it stays almost constant for higher admixtures. This is again more an effect of the slow gas flow velocity as the density that reaches the flow cell is quite low, so no significant change can be observed. Admixtures higher than 0.5% even dip a little in their O₂(a¹Δ_g) density. Here, the plug flow simulation predicted higher ozone densities than 2D fluid simulation and experiment, as seen in figure 8, which led to higher destruction rates of O₂(a¹Δ_g).

The plug flow simulation shows a very good agreement with the absolute experimental values, although it falls short in describing the trends correctly due to the likely underestimated gas flow velocity in the flow cell region. The 2D fluid simulation matched the experimental trends very well, but overestimates the O₂(a¹Δ_g) densities. This is attributed to the limited computation time of the simulation, which does not reach an equilibrium state. The values could potentially come very close to the experiment with longer simulation time, as shown for high flows, which have a better agreement with the experiment.

4.3. O₂(b¹Σ_g⁺) density

The O₂(b¹Σ_g⁺) spectra were taken with an integration time of 1 s. The O₂(b¹Σ_g⁺) density in the jet was stable directly after plasma ignition if the jet had already heated up. Because the O₂(b¹Σ_g⁺) is measured inside the COST jet without flow cell, no comparison to the 2D fluid simulation was conducted.

Figure 14 compares the O₂(b¹Σ_g⁺) density distribution along the COST jet discharge channel for the plug flow simulation and the experimental results. After an initial rise, the measured O₂(b¹Σ_g⁺) density decreases slowly while the simulation shows a maximum at approx. -17 mm in the discharge channel. Because the density values are heavily dependent on the position inside the jet and the oxygen admixture, as shown in figure 15, we concentrated on this parameter to illustrate the differences between plug flow simulation and experimentally determined values.

Figure 15 shows the measured O₂(b¹Σ_g⁺) density distribution along the discharge channel for varying admixtures of oxygen in comparison with the 1D plug flow model. For an admixture of 0.1% of oxygen, the measured O₂(b¹Σ_g⁺) density increases asymptotically and reaches its maximum at the end of the discharge channel, before decaying in the effluent. For an admixture of 0.3% the density increases slightly, and the position of the maximum is already reached at approx. -8 mm instead at the jets' exit. Afterwards, it decreases slowly. The same behaviour can be observed for higher admixtures. The maximum position shifts towards the gas inlet of the discharge channel, and

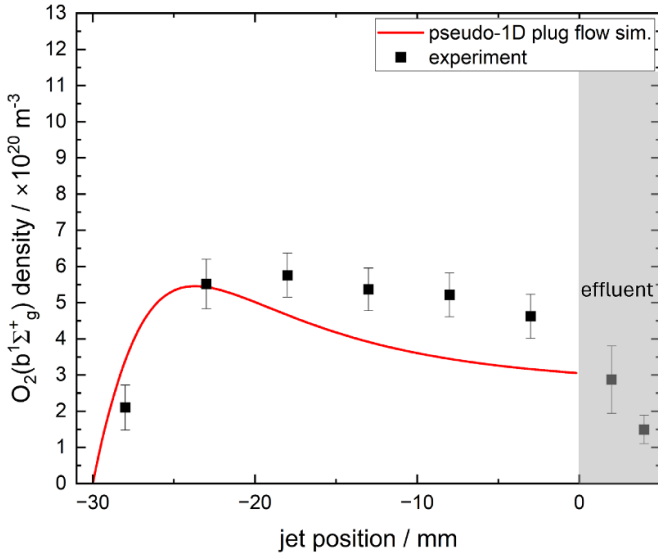


Figure 14. $O_2(b^1\Sigma_g^+)$ density distribution comparison between simulation and experiment. Grey area indicates effluent region. Example for a plasma power of 1 W, 1 slm He flow, and a 0.5% oxygen admixture as a function of the discharge channel position in the jet.

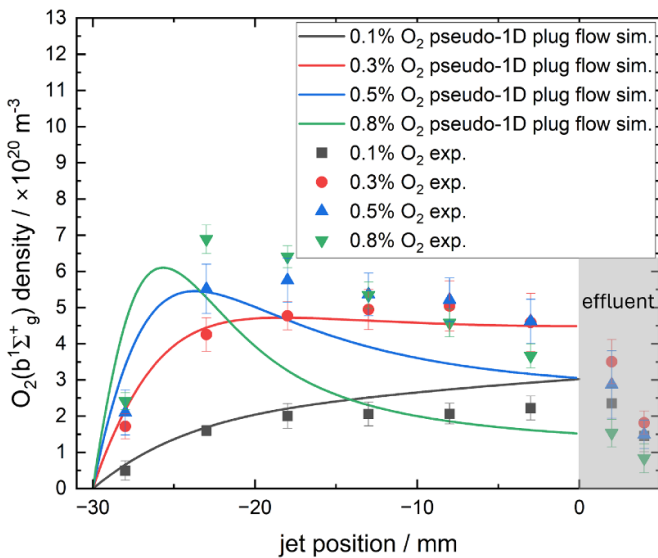


Figure 15. $O_2(b^1\Sigma_g^+)$ density distribution comparison along the gas channel between simulation and experiment. Grey area indicates effluent region. Example for a power of 1 W and 1 slm He flow and an oxygen admixture variation.

the maximum density increases slightly with higher oxygen admixture.

The simulation follows the experimental values very well for 0.1% and 0.3% oxygen admixture. For higher admixtures, a maximum in the first third of the discharge can be observed. Its position gets closer to the start of the jet with increasing admixture as observed in the experiment. The density maximum also increases with the oxygen admixture. The

decay after reaching the maximum is faster than in the experimental case, which leads to lower densities at the end of the discharge channel compared to the experiment.

A possible explanation for this behaviour is the overestimation of the ozone density by the simulation (compare e.g. figure 6). The ozone density increases along the discharge channel to the exit of the jet, leading to higher consumption of $O_2(b^1\Sigma_g^+)$. As the ozone density is higher when closer to the end of the discharge, the simulated $O_2(b^1\Sigma_g^+)$ density reaches its maximum at the beginning of the discharge channel and decays quickly towards the end. This is consistent with the ozone density of the simulation being overestimated, especially with the rising oxygen admixture as shown in figure 8.

In the COST jet, the $O_2(b^1\Sigma_g^+)$ density predicted by the plug flow simulation has a very good agreement with the experimental values and depicts the experimental trends quite well. Higher admixtures, however, lead to a discrepancy in the consumption rate of $O_2(b^1\Sigma_g^+)$. The overestimated consumption rate is likely caused by the overestimated ozone density, as shown in the figures 6–8. These observations agree with the ozone measurements and confirm the suggested use cases of the two simulations.

5. Conclusion

The aim of this work was the measurement of the excited molecular oxygen species $O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma_g^+)$ produced within the COST atmospheric pressure plasma jet, supplied with a helium-oxygen gas mixture. The experimental results were compared to two simulations to validate the measured values and also to investigate established COST jet models for their applicability to the effluent region.

Comparison of the experimental results to the pseudo-1D plug flow simulation as well as the 2D fluid simulation showed that both simulations were capable of computing the species' densities in agreement with the experiment, as both are within the same order of magnitude as the experimental densities. This makes the plug flow simulation an effective tool to compute a good estimate of the expected densities even for complicated effluent geometries. The 2D fluid simulation is able to accurately describe trends of the reactive species' densities for power, flow, and admixture variation. It is therefore well suited if the density distribution of reactive species in the effluent is critical or of special interest. Employing longer computation times to reach an equilibrium state within the flow cell might further increase the agreement with the experiment. The simulations overestimate the ozone density for oxygen admixtures higher than 0.1% as shown in the ozone density section. This was confirmed by the $O_2(b^1\Sigma_g^+)$ measurements as the consumption rate of $O_2(b^1\Sigma_g^+)$ towards the jet exit was higher when compared to the experimental values. This points to a potential area of weakness in the reaction scheme used within the simulations, which might also explain discrepancies to the measured densities in the flow cell. This is consistent with the conclusions of the work of Turner [34],

who described the close coupling between the densities of $O_2(b^1\Sigma_g^+)$ and ozone under similar conditions to those studied here. Additionally, the wall reactions for the oxygen species used in the simulations are not well known under the conditions used in this work. In reality, these may also deviate between the plasma and effluent regions and are even excluded for ozone. As a result, further work on improving the description of wall reactions may also improve agreement between experiment and simulation.

Both quantitative and qualitative differences were observed between the results of the plug flow and 2D fluid simulations. While differences in the treatment of residence time and the direct simulation of the spatial profiles of reactive species in the 2D fluid simulations are likely to be a key reason for these, the different reaction schemes used in both simulations are also likely to be a factor. In general, the reaction schemes used in each simulation are similar, but not identical. Given the sensitivity of plasma-chemical simulations to the details of certain individual reactions and the species included, even small differences can be expected to lead to noticeable changes in reactive species densities even when the physical model is unchanged, as described in [34], for example.

To enhance the agreement of the experimental results with the simulations, one could improve on the flow cell design. Reducing the appearance of vortices might reduce the simulation time for fluid models and might also lessen the discrepancy in the flow velocity considered in the plug flow simulation. Furthermore, some of the less validated rate coefficients for various reactions, particularly wall interactions, could be improved by taking a more differentiated look at the plasma and effluent region, and carefully varying those coefficients in the simulations. Nevertheless, the employed simulations showed that even outside their ‘comfort zone’, they lead to a reasonably good agreement, with the experiment highlighting the quality of the plasma chemistry validation.

As atmospheric pressure plasma jets are mostly used without confining the effluent, it is important to know to which extent the measured densities can be applied to unconfined effluent conditions. Because of the relatively long lifetimes of the investigated species, notably $O_2(a^1\Delta_g)$ and ozone, it is reasonable to assume that the densities are very much transferable to a non-confined effluent. It should be noted, however, that this holds true mostly for laminar flows. However, gas dynamics might deviate drastically between different application geometries. Mixing of gases due to turbulent conditions can therefore have a high impact on the measured densities. Although, at least when considering the absolute density magnitudes, this study should serve as a good base of comparison and highlights the importance of taking the high interaction of these species into account.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: [https://rdpcidat.rub.de/\[64\]](https://rdpcidat.rub.de/[64]).

Acknowledgments

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Author contributions

Sascha Chur  0000-0001-6926-3747

Conceptualization (lead), Methodology (lead), Formal analysis (lead), Investigation (lead), Data curation (lead), Writing – original draft (lead), Visualization (lead), Supervision (lead)

Robin Minke

Methodology (lead), Validation (lead), Formal analysis (lead), Investigation (lead)

Youfan He  0000-0003-1275-7695

Methodology (lead), Software (lead), Validation (lead), Formal analysis (lead), Writing – original draft (lead)

Máté Vass  0000-0001-9865-4982

Methodology (lead), Software (lead), Validation (lead), Formal analysis (lead), Writing – original draft (lead), Visualization (lead)

Thomas Mussenbrock

Resources (lead), Supervision (lead), Project administration (lead)

Ralf Peter Brinkmann

Resources (lead), Supervision (lead), Project administration (lead)

Efe Kemaneci  0000-0002-5540-0947

Resources (lead), Supervision (lead), Project administration (lead)

Lars Schücke

Writing – review & editing (lead), Supervision (lead), Project administration (lead)

Volker Schulz-von der Gathen  0000-0002-7182-3253

Writing – review & editing (lead), Supervision (lead)

Andrew R Gibson  0000-0002-1082-4359

Methodology (supporting), Writing – review & editing (supporting), Supervision (supporting), Project administration (supporting)

Marc Böke  0000-0003-1062-5808

Writing – review & editing (lead), Resources (lead), Supervision (lead), Project administration (lead)

Judith Golda  0000-0003-2344-2146

Writing – review & editing (lead), Resources (lead), Supervision (lead), Project administration (lead)

Appendix

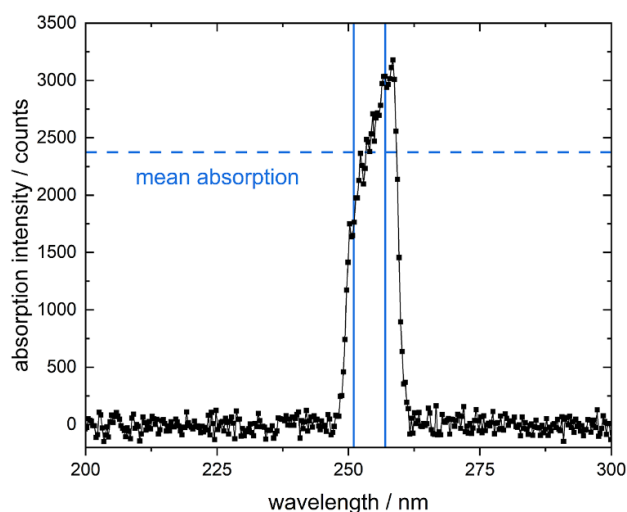


Figure 16. Absorption measurement of the ozone spectra.

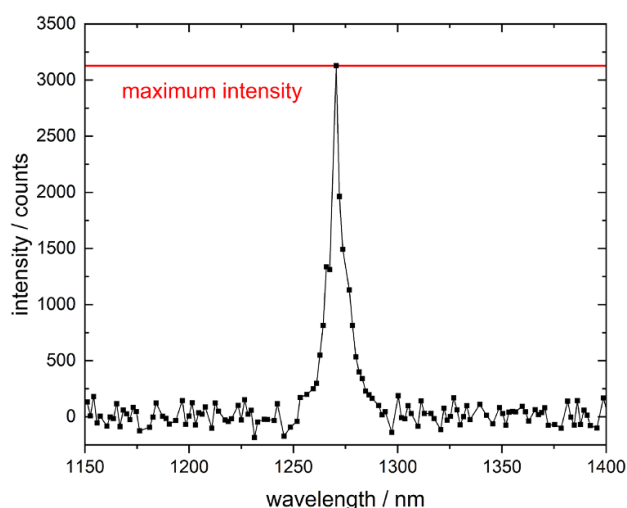


Figure 17. Intensity measurement for the $O_2(a^1\Delta_g)$ spectra.

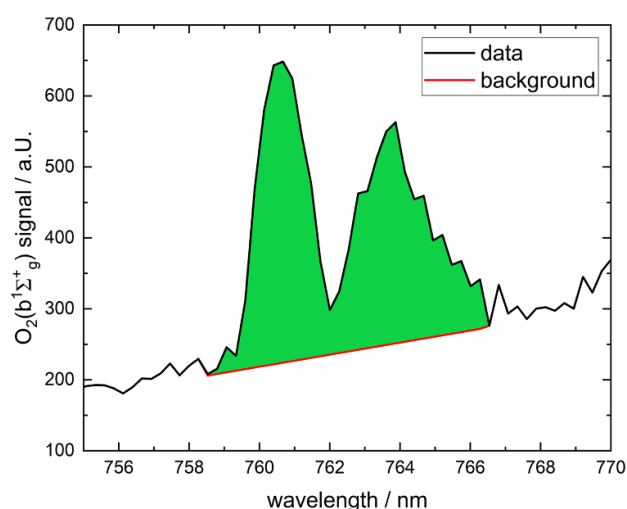


Figure 18. Intensity measurement of the $O_2(b^1\Sigma_g^+)$ molecular spectrum.

References

- [1] Lu X, Laroussi M and Puech V 2012 *Plasma Sources Sci. Technol.* **21** 034005
- [2] Winter J, Brandenburg R and Weltmann K D 2015 *Plasma Sources Sci. Technol.* **24** 064001
- [3] Robert E, Sarron V, Riès D, Dozias S, Vandamme M and Pouvesle J M 2012 *Plasma Sources Sci. Technol.* **21** 034017
- [4] Sobota A, Guaitella O, Sretenović G B, Krstić I B, Kovačević V V, Obrusnik A, Nguyen Y N, Zajičková L, Obradović B M and Kuraica M M 2016 *Plasma Sources Sci. Technol.* **25** 065026
- [5] Golda J et al 2016 *J. Phys. D: Appl. Phys.* **49** 084003
- [6] Reuter S, Von Woedtke T and Weltmann K D 2018 *J. Phys. D: Appl. Phys.* **51** 233001
- [7] Pawlat J, Terebun P, Kwiatkowski M and Diatczyk J 2016 *J. Phys. D: Appl. Phys.* **49** 374001
- [8] Shaw D, West A, Bredin J and Wagenaars E 2016 *Plasma Sources Sci. Technol.* **25** 065018
- [9] Ichiki T, Taura R and Horiike Y 2004 *J. Appl. Phys.* **95** 35–39
- [10] Benedikt J, Raballand V, Yanguas-Gil A, Focke K and Von Keudell A 2007 *Plasma Phys. Control. Fusion* **49** B419–27
- [11] Waskoenig J, Niemi K, Knake N, Graham L M, Reuter S, Gathen V S v D and Gans T 2010 *Plasma Sources Sci. Technol.* **19** 045018
- [12] Preissing P, Korolov I, Schulze J, Schulz-von Der Gathen V and Böke M 2020 *Plasma Sources Sci. Technol.* **29** 125001
- [13] Bischoff L et al 2018 *Plasma Sources Sci. Technol.* **27** 125009
- [14] Maletić D, Puač N, Lazović S, Malović G, Gans T, Schulz-von Der Gathen V and Petrović Z L 2012 *Plasma Phys. Control. Fusion* **54** 124046
- [15] Liu Y, Korolov I, Trieschmann J, Steuer D, Schulz-von Der Gathen V, Böke M, Bischoff L, Hübner G, Schulze J and Mussenbrock T 2021 *Plasma Sources Sci. Technol.* **30** 064001
- [16] Korolov I, Steuer D, Bischoff L, Hübner G, Liu Y, Schulz-von Der Gathen V, Böke M, Mussenbrock T and Schulze J 2021 *J. Phys. D: Appl. Phys.* **54** 125203
- [17] Graves D B 2014 *Plasma Process. Polym.* **11** 1120–7
- [18] Yang H, Villani R M, Wang H, Simpson M J, Roberts M S, Tang M and Liang X 2018 *J. Exp. Clin. Cancer Res.* **37** 266
- [19] Bekeschus S, Von Woedtke T, Emmert S and Schmidt A 2021 *Redox Biol.* **46** 102116
- [20] Kong M G, Kroesen G, Morfill G, Nosenko T, Shimizu T, Van Dijk J and Zimmermann J L 2009 *New J. Phys.* **11** 115012
- [21] Sousa J S, Niemi K, Cox L J, Algwari Q T, Gans T and O'Connell D 2011 *J. Appl. Phys.* **109** 123302
- [22] Jablonowski H, Santos Sousa J, Weltmann K D, Wende K and Reuter S 2018 *Sci. Rep.* **8** 12195
- [23] Lazzaroni C, Chabert P, Lieberman M A, Lichtenberg A J and Leblanc A 2012 *Plasma Sources Sci. Technol.* **21** 035013
- [24] Liu D X, Bruggeman P, Iza F, Rong M Z and Kong M G 2010 *Plasma Sources Sci. Technol.* **19** 025018
- [25] Schröter S et al 2018 *Phys. Chem. Chem. Phys.* **20** 24263–86
- [26] Schröter S, Gibson A R, Kushner M J, Gans T and O'Connell D 2018 *Plasma Phys. Control. Fusion* **60** 014035
- [27] Murakami T, Niemi K, Gans T, O'Connell D and Graham W G 2014 *Plasma Sources Sci. Technol.* **23** 025005
- [28] Murakami T, Niemi K, Gans T, O'Connell D and Graham W G 2013 *Plasma Sources Sci. Technol.* **22** 045010
- [29] Murakami T, Niemi K, Gans T, O'Connell D and Graham W G 2012 *Plasma Sources Sci. Technol.* **22** 015003
- [30] Sun B, Liu D, Iza F, Wang S, Yang A, Liu Z, Rong M and Wang X 2019 *Plasma Sources Sci. Technol.* **28** 035006

- [31] Gaens W V and Bogaerts A 2013 *J. Phys. D: Appl. Phys.* **46** 275201
- [32] Gaens W V and Bogaerts A 2014 *Plasma Sources Sci. Technol.* **23** 035015
- [33] Gaens W V, Bruggeman P J and Bogaerts A 2014 *New J. Phys.* **16** 063054
- [34] Turner M M 2015 *Plasma Sources Sci. Technol.* **24** 035027
- [35] Liu D, Sun B, Iza F, Xu D, Wang X, Rong M and Kong M G 2017 *Plasma Sources Sci. Technol.* **26** 045009
- [36] Schmidt-Bleker A, Winter J, Bösel A, Reuter S and Weltmann K D 2016 *Plasma Sources Sci. Technol.* **25** 015005
- [37] Schmidt-Bleker A, Winter J, Iseni S, Dünnebier M, Weltmann K D and Reuter S 2014 *J. Phys. D: Appl. Phys.* **47** 145201
- [38] Stafford D S and Kushner M J 2004 *J. Appl. Phys.* **96** 2451–65
- [39] Liu Y, Liu D, Zhang J, Sun B, Yang A and Kong M G 2020 *Phys. Plasmas* **27** 043512
- [40] Kelly S, Golda J, Turner M M and Schulz-von Der Gathen V 2015 *J. Phys. D: Appl. Phys.* **48** 444002
- [41] Viegas P, Slikboer E, Bonaventura Z, Guaitella O, Sobota A and Bourdon A 2022 *Plasma Sources Sci. Technol.* **31** 053001
- [42] Vass M, Schulenberg D, Donkó Z, Korolov I, Hartmann P, Schulze J and Mussenbrock T 2024 *Plasma Sources Sci. Technol.* **33** 015012
- [43] Klich M, Wilczek S, Donkó Z and Brinkmann R P 2022 *Plasma Sources Sci. Technol.* **31** 045003
- [44] Hemke T, Wollny A, Gebhardt M, Brinkmann R P and Mussenbrock T 2011 *J. Phys. D: Appl. Phys.* **44** 285206
- [45] Golda J, Held J and Gathen V S v D 2020 *Plasma Sources Sci. Technol.* **29** 025014
- [46] He Y et al 2021 *Plasma Sources Sci. Technol.* **30** 105017
- [47] Booth J P et al 2022 *Plasma Sources Sci. Technol.* **31** 065012
- [48] Ionin A A, Kochetov I V, Napartovich A P and Yuryshev N N 2007 *J. Phys. D: Appl. Phys.* **40** R25–R61
- [49] Sousa J S, Bauville G, Lacour B, Puech V, Touzeau M and Pitchford L C 2008 *Appl. Phys. Lett.* **93** 011502
- [50] Niermann B, Böke M, Sadeghi N and Winter J 2010 *Eur. Phys. J. D* **60** 489–95
- [51] Schulz von Der Gathen V, Buck V, Gans T, Knake N, Niemi K, Reuter S, Schaper L and Winter J 2007 *Contrib. Plasma Phys.* **47** 510–9
- [52] Winter J, Dünnebier M, Schmidt-Bleker A, Meshchanov A, Reuter S and Weltmann K D 2012 *J. Phys. D: Appl. Phys.* **45** 385201
- [53] Inn E C Y and Tanaka Y 1953 *J. Opt. Soc. Am.* **43** 870010
- [54] Inoue Y and Ono R 2017 *J. Phys. D: Appl. Phys.* **50** 214001
- [55] He Y, Brinkmann R P, Kemaneci E and Gibson A R 2025 *Plasma Sources Sci. Technol.* (submitted) (<https://doi.org/10.1088/1361-6595/adffe6>)
- [56] Chantray P J 1987 *J. Appl. Phys.* **62** 1141–8
- [57] del Caz A T, Guerra V, Gonçalves D, da Silva M L, Marques L, Pinhão N, Pintassilgo C D and Alves L L 2019 *Plasma Sources Sci. Technol.* **28** 043001
- [58] Kushner M J 2004 *J. Appl. Phys.* **95** 846–59
- [59] Kushner M J 2005 *J. Phys. D: Appl. Phys.* **38** 1633–43
- [60] Liu Y et al 2023 *Plasma Sources Sci. Technol.* **32** 025012
- [61] Steuer D, Korolov I, Chur S, Schulze J, Schulz-von Der Gathen V, Golda J and Böke M 2021 *J. Phys. D: Appl. Phys.* **54** 355204
- [62] Wijaikhum A, Schröder D, Schröter S, Gibson A R, Niemi K, Friderich J, Greb A, Schulz-von Der Gathen V, O'Connell D and Gans T 2017 *Plasma Sources Sci. Technol.* **26** 115004
- [63] Bang S, Snoeckx R and Cha M S 2023 *Plasma Chem. Plasma Process.* **43** 1453–72
- [64] Sascha C 2025 Dynamics of reactive oxygen species produced by the COST microplasma jet, Research Data Repository Research Department PLasmas with Complex Interactions (available at: <https://rdpcidat.rub.de/>)