Generation of Singlet Oxygen in HV Pulsed + DC Crossed Discharge at Atmospheric Pressure for Oxygen-Enhanced Combustion

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Abstract—Promoting action of electric discharges on fuel combustion processes results in reduction of the ignition delay, improvement of the flame stability as well as the extension of the flammability limits and these are key technical issues in combustion improvement [1, 2]. One promising approach consists in plasma enhanced activation of oxidizing substance, e.g. molecular oxygen to its electronically excited singlet delta O2(a1Δ) and singlet sigma O2(b1Σ) states. In contrast with non-excited reactants, singlet oxygen molecules are more chemically active and can affect reaction kinetics due to decrease of the barrier of endoenergetic reactions. The present work presents results of our recent feasibility experiments on singlet oxygen generation in non-equilibrium electric discharges, from reduced pressures up to atmospheric pressure and for different discharge powers. A high voltage pulse generator (PPS) capable of delivering 50 ns electric pulses of 20 kV with a controlled voltage rise time of 20 ns at a frequency up to 25 kHz has been used. We are able to produce some quantities of O2(b1Σ) molecules and detect them by optical emission spectroscopy in the visible band O2(b1Σ→X1Σ) at 762 nm. Singlet oxygen generation has been performed in O2/He binary mixtures in the application of self-sustained and non-self-sustained discharges.

Keywords—Plasma-assisted-combustion, excited oxygen, O2(b1Σ), O2(a1Δ), dielectric barrier discharge, high voltage pulse generator

I. INTRODUCTION

The present study is prompted by the ever-increasing need to reduce emissions from combustors to satisfy the legislation which is more and more drastic. It is of vital interest to maximize the efficiency of various combustion processes to save fuel. The object of this work is to significantly improve combustion and reduce pollutant emissions with plasma oxidiser “activation”. At present, relatively little is known about the possible effects of electronically excited species on combustion kinetics. However, if present in critical concentrations, they may reduce ignition delay time [2], because typical recombination / relaxation energies tend to be very high (of order 10 eV against 1 eV for recombination of radicals). In addition to combustion applications, singlet delta oxygen is also important as an energy source for lasers, the chemical oxygen iodine laser (COIL), and the discharge oxygen iodine laser (DOIL). In these lasers, the excitation of the singlet delta oxygen is transferred to iodine molecules which are then the active lasing medium [3, 4 and 8]. Preliminary numerical modeling has shown that the transition to a non-self-sustained mode can significantly improve the efficiency of singlet oxygen generation in the discharge. For that, an original double discharge plasma-chemical reactor has been developed at Heat and Mass Transfer Institute (HTMI, Minsk). In this crossed discharge, short high voltage DBD pulses produce ionization while a comparatively low electric DC field supports the electric current between ionizing pulses. This allows adjusting the electric field close to the optimum for excitation of singlet oxygen molecules to increase the production yield. Optimization of discharge parameters and modification of the discharge cell design in contrast with [3] allow us to operate at elevated pressures (up to atmospheric) and enlarge specific electric energy input (up to 10 eV/molecules) while keeping good discharge uniformity. This investigation is devoted to the study of O2(a1Δ) and O2(b1Σg+) production in binary mixtures of oxygen and helium. We used a simple dielectric barrier discharge and a crossed discharge fed by a high voltage pulsed generator with particular characteristics. Emission spectroscopy has been employed to characterise these active species in the discharge and their variations with experimental parameters (discharge current and voltage, pressure, deposited power, gases flows…). The densities of oxygen molecules excited in the metastable states O2(a1Δ) and O2(b1Σg+) is determined on intensity of the band O2(a1Δ, v=0) → O2(X1Σg) at 1268 nm and O2(b1Σg+, v=0) → O2(X1Σg) at 762 nm.

II. EXPERIMENTAL

A. Simple dielectric barrier discharge
Experiments are conducted at a new plasma combustion facility recently developed at ICARE Institute. It consists in a stainless steel low pressure tank housing the discharge cell. A premixed helium/oxygen flow is delivered to the DBD section via a supply line. A schematic of the DBD used in this study is presented in fig. 1:

![Figure 1. Discharge cell.](image)

The DBD is composed of two parallel copper-based electrodes bounding a 4 mm gas space, covered with a 1 mm thick ceramic plate. It is worth noting that the maximal temperature use for these electrodes is 250°C. The characteristic resides in its strong dielectric permittivity ($\varepsilon > 1000$). The pulsed electrodes are powered by a high voltage pulsed generator (20 kV and 25 kHz) specially constructed at HMTI. When the applied voltage exceeds the breakdown voltage, electron avalanches occur, resulting in the formation of a forest of micro streamers.

The plasma was operated with oxygen and helium or argon gas mixture at 50 Torr total pressure up to atmospheric pressure, and for various flow velocities. The tank is pumped out using a primary pump. The gases used were specified to be 99.999% pure. Considerable care is taken to ensure cell cleanliness and O$_2$ purity. The experimental conditions (mainly the applied voltage and electrode structure) were controlled so that no arcing was observed. It is worth noting that electric discharge sustained in moderate reduced pressures of oxygen is prone to arcing. The characteristics of the alimentation facility are presented in table 1; the FWHM depends on the discharge characteristics:

<table>
<thead>
<tr>
<th>$U_{\text{min}}$ (kV)</th>
<th>FWHM (ns)</th>
<th>$U_{\text{rise}}$ time (ns)</th>
<th>Frequency (kHz)</th>
<th>Polarity</th>
<th>Discharge gap (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>50</td>
<td>20</td>
<td>0.001 - 25</td>
<td>$&gt;0$ or $&lt;0$</td>
<td>4</td>
</tr>
</tbody>
</table>

Under the conditions mentioned above, the average value of reduced electric field, defined as $E/N$, is strictly superior to 30 Townsend (Td), where $E = U/d$ is the spatial average of the electric field strength and $n$ is the particles’ number density.

The experimental difficulties of these discharges are obvious: this nanosecond pulsed discharge requires on the one hand, equipment with nanosecond temporal resolution, and on the other hand, they generate a high electromagnetic noise [7]. Several works underline that for pulsed discharges, there is an electromagnetic compatibility (EMC) problem of the high voltage source with the equipment around [3, 6, 7]. Then, in our experimental set-up, to prevent interference of the high voltage pulse generator with the diagnostic equipment, the entire test section assembly (HVPG) is shielded by a copper Faraday cage and connections by ferrites and adapted and filtered connections.

### B. Crossed discharge plasma reactor

The measurements with a crossed-discharge reactor (CDR) have been conducted at HMTI, Minsk. Binary mixtures are mixed up in a T-typed mixer at atmospheric pressure and flows to the CDR with a rectangular section of 6.5x16 mm$^2$. The general flow rate of a mixture varied from 0.5 to 10 l/min. Concentration of oxygen in the mixture varied from 0.5 % to 5 %. Body of CDR is executed from glass ceramics. Both electrodes (36x24 mm$^2$) of the pulse barrier discharge (Pulse DBD electrodes) are covered by dielectric of 1 mm thickness; the gas gap (Pulse gap) is 6.5 mm. Thickness of pulse electrodes is 0.15 mm. The pulse barrier discharge electrodes were connected to the HVPG. Open DC electrodes have been executed from stainless steel; their sizes are 40x6.5 mm$^2$ and 20x6.5 mm$^2$ accordingly. Edges of open electrodes were rounded with curving radius of about 3.2 mm. Also their surfaces were polished and are exposed to the flow. Open electrodes were connected to HV DCPS (high voltage direct current power supply) through matched high-voltage chokes and ballast resistance. The minimal distance between DC electrodes (DC gap) was 16 mm. The pulse barrier discharge of preionization was lit from HVPG. Original HVPG has adjustable voltage (from 0 to 20 kV) and pulse frequency ranging from 1 Hz to 25 kHz, with pulse duration at half-height of about 50 nanoseconds (Fig. 3). The barrier discharge reactor is nonlinear load; HVPG is matched with this load. Between high-voltage pulses, the discharge was sustained by the HV DCPS. The voltage was adjusted from 0 up to 2 kV, DC current changed from 0 to 100 mA (Fig. 3) and ballast resistance changed in experiments from hundreds Ohms up to 10 kΩ. The reactor with fiber optics sensors linked to the spectrometers permits the optical diagnostics.

A schematic of experimental set-up as well as a 3D design of the crossed discharge reactor (CDR) are presented in fig. 2 left and right respectively:

![Figure 2. Experimental set-up (left) and crossed discharge cell cross section (right) for singlet oxygen generation. Flow is left to right](image)
B. Optical emission spectroscopy (OES)

Optical emission spectroscopy (OES) is used for diagnostics of reactive plasmas. The main advantage of the method is the non-invasive character of measurements.

Fig. 4. Diagram of apparatus used for optical emission spectroscopy in the visible range

B.1 \(\text{O}_2(\text{b}^1\Sigma^+)\) emission band at 762 nm

The plasma combustion facility is designed for the use of spectroscopic diagnostics. In particular, the low pressure tank has two optical access CaF\(_2\) windows, as shown in fig. 4. A SOPRA F1500 monochromator with a grating of 1800 grooves/mm is used to select the desired wavelength region. The angle of the grating is changed directly mounted to the exit of the monochromator for optical multi-channel array (OMA) of 1024 pixels is shown in fig. 4. A SOPRA F1500 monochromator with a grating of 1800 grooves/mm is used to select the desired wavelength region. The angle of the grating is changed to modify the position of the rotational energy levels related to the quantum number of spin \(S\). The expression of the rotational energy levels is different according to the spin degeneration: doublet (\(S=1/2\), triplet (\(S=1\)) states, but also the electronic quantum number \(\Lambda\) (states \(\Sigma, \Pi, \Delta\ldots\)) is a constant for a given electronic state. The various additional interactions resulting from these couplings are then introduced in the form of additional spectroscopic constants: the spin-orbit interactions are expressed by the spectroscopic constant \(A_s\); the spin-spin interactions are expressed by the spectroscopic constant \(\gamma_s\); and the spin-rotation interactions are expressed by the spectroscopic constant \(\gamma_r\). For a singlet sigma state, \(\Sigma\) (for \(\text{b}^1\Sigma^+\)), rotational terms is written: \(F(J)=B_s(J(J+1))-D_s(J(J+1))^2-H_s(J(J+1))^3\), where \(B_s\) is the rotational constant and \(D_s\) and \(H_s\) are the quartic centrifugal distortion and the sextic centrifugal distortion constants respectively [12]. For a triplet splitting of the \(\text{b}^3\Sigma^+\) ground state of the \(\text{O}_2\) molecule (\(\Sigma^1\Sigma^+\)), theoretical expressions for the rotational energy: \(F_r(J), F_T(J)\) and \(F_s(J)\) are given by reference [12]. Finally, in order to calculate the rotational line intensities, theoretical Hönzl-London factors are integrated in the program.

B.2 \(\text{O}_2(a^1\Delta_g)\) emission band at 1.27 \(\mu\)m

Singlet delta oxygen emits in the infrared between 1.2 and 1.3 \(\mu\)m. To record the spectrum, a liquid nitrogen cooled InSb detector was employed with the monochromator. The signal from the InSb detector was passed through a computer for data acquisition. To eliminate the background, a lock-in amplifier was utilized with an optical chopper, which was mounted at the entrance of the spectrometer.

III. SPECTRA SIMULATION

The program PGOPHER\(^\text{\texttrademark}\) was used and compared to our simulation program for both transitions \(\text{O}_2 (\text{a}^1\Delta_g) \rightarrow \text{O}_2 (X^3\Sigma_g^+) (0,0)\) (infrared band) and \(\text{O}_2 (\text{b}^1\Sigma_g^+) \rightarrow \text{O}_2 (X^3\Sigma_g^+) (0,0)\) (atmospheric ‘\(\text{A}\)’ band) of molecular oxygen. Briefly, PGOPHER\(^\text{\texttrademark}\) is a general purpose program for simulating and fitting rotational spectra [9]. To perform a simulation, we have to create an input file containing the rotational constants and other settings required.

A. Atmospheric ‘\(\text{A}\)’ oxygen band

The oxygen atmospheric ‘\(\text{A}\)’ band, corresponding to the (0,0) band of the \(\text{b}^1\Sigma_g^+ \rightarrow \text{X}^3\Sigma_g^+\) system is a prominent feature in the absorption and emission spectrum of the terrestrial atmosphere [11]. Nevertheless, these bands are very weak due to the strongly forbidden character of gerade-gerade and \(\Sigma^-\Sigma^+\) transitions, and it can only be observed via a magnetic dipole transition moment. The coupling phenomena between the movement of electrons and the rotational movement results in modifying the position of the rotational energy levels related to the quantum number of spin \(N\). The expression of the rotational energy levels is different according to the spin degeneration: doublet (\(S=1/2\)), triplet (\(S=1\)) states...
B. Infrared oxygen band

The O$_2$ $^1\Delta_g \rightarrow ^3\Sigma^+$ transition is electric-dipole forbidden, but can be induced by magnetic-dipole and electric quadrupole interactions. The theoretical expression for the rotational energy of $^1\Delta_g$ ($v=0$) state is written:

$$F(J) = v_0 + B_0(J(J+1) - A_0 - 1)^2,$$

with $\Lambda=2$ appropriate for this state.

Molecular constants for $b^1\Sigma^+$ state of $^{16}$O$_2$ of Babcock and Herzberg [11, 12] were employed. For $a^1\Delta_g$ state, those of Western [9] were used. It is worth noting that molecular constants obtained by other workers were used for comparisons.

Fig. 5 shows the oxygen ‘A’ band and the infrared oxygen band spectra. A comparison between simulated spectra performed by the program PGOPHER® and using our simulation program is shown.

In our simulation program, each line was fitted according to a Gaussian shape. In the PGOPHER® program, lines were fitted with a variable combination of Gaussian and Lorentzian profiles.

One can observe a good fitting in line positions between both simulation programs. The differences between spectra at the basis would come from this fitting difference (Lorentzian line shape to account for pressure broadening in the PGOPHER® program).

The characteristic structure of the red atmospheric oxygen “A” band is a R-form branch forming a head and a P-form branch, separated from the former by a zero gap. The singlet delta emission, O$_2$(a$^1\Delta_g$) to O$_2$(X$^3\Sigma^+_g$) lets clearly appear the P, Q, and R branches at 1264, 1269, and 1274 nm.

IV. RESULTS AND DISCUSSIONS

A. Numerical simulation

An important parameter for the discharge is the value of the reduced electric field (which is directly connected to the electronic temperature, $(E/N)$, usually expressed in Townsend varies from 10 to 100 Td for a DBD at atmospheric pressure.

The following calculations are performed by a Boltzmann solver Bolsig$^\text{®}$ [10] with a set of cross sections used as inputs. The Boltzmann solver calculates the energy electron distribution function in the plasma as well as the gas dissociation and the electronic excited states population, and provides the resultant electron impact process rate coefficients as a function of $(E/N)$.

![Fig. 6. Cross sections for electronic excitation from the ground state: left, Excitation of $a^1\Delta_g$ ($E_{exc}=0.98$ eV); right, excitation of $b^1\Sigma^+$ ($E_{exc}=1.63$ eV)].

Figure 7 shows the dependence of specific energy deposited to excite O$_2$ in its first and second singlet states ($O_2(a^1\Delta_g)$ and $O_2(b^1\Sigma^+_g)$), to excite the vibrations ($v=1$) and for O$_2$ dissociation, according to $(E/N)$ and for O$_2$/He mixtures:

![Fig. 7. Fractional energy losses of electrons in pure O$_2$ against reduced electric field $(E/N)$ for excitation of the vibrations $v=1$ $(E_{exc}=0.19eV)$, excitation of $a^1\Delta_g$ $(E_{exc}=0.98eV)$, excitation of $b^1\Sigma^+$ $(E_{exc}=1.63eV)$ and O$_2$ dissociation $(E_{exc}=8.4eV)$].

For pure O$_2$, over 45% of energy goes to excitation of O$_2(a^1\Delta_g)$ state and over 15% goes to excitation of O$_2(b^1\Sigma^+_g)$ state. One can observe from these calculations that the energy fraction going into electron impact excitation of O$_2(a^1\Delta_g)$ and O$_2(b^1\Sigma^+_g)$ states in pure O$_2$ mixture reaches a maximum at ~ 10 Td. This is the optimal value of energy deposited for an optimal singlet oxygen excitation.

The following graphs show the dependence of specific energy deposited to excite O$_2$ in its first and second singlet state with the reduced electric field $(E/N)$.

![Fig. 8. Fractional energy losses of electrons in pure O$_2$ against reduced electric field $(E/N)$ for excitation of $a^1\Delta_g$ $(E_{exc}=0.98eV)$ (left), and $b^1\Sigma^+$ $(E_{exc}=1.63eV)$ (right) and for different percentages of O$_2$ in binary mixtures O$_2$/He]
One can observe from fig. 8 that the maximal efficiency of \( \text{O}_2(a^1 \Delta_g) \) and \( \text{O}_2(b^1 \Sigma_g^+) \) excitation decreases with He diluting. In the same time, the optimal \((E/N)\) corresponding to the maximal singlet oxygen excitation decreases with He diluting (10 Td, 7.5 Td and 5 Td for pure \( \text{O}_2 \), 50% \( \text{O}_2 \) and 10% \( \text{O}_2 \) diluted in helium respectively).

It is worth noting that this \((E/N)\) value is considerably lower than those achieved in self-sustained nonequilibrium electric discharge. These results are in agreement with literature results [2, 3, 5, 6, and 7]. Indeed, recent investigations on electric discharge production of \( \text{O}_2(a^1 \Delta_g) \) consisted on engineering the \((E/N)\) nearer to the optimum value for \( \text{O}_2(a^1 \Delta_g) \) production. In pure oxygen, the optimal value of \( \sim 10 \text{Td} \) is obtained.

To conclude, in order to optimize the \( \text{O}_2(b^1 \Sigma_g^+) \) and \( \text{O}_2(a^1 \Delta_g) \) yield in the plasma, the discharge should operate at \((E/N)\) values where the energy input into the target states is maximum. These preliminary numerical modelling has shown that the transition to a non-self-sustained mode can significantly improve the efficiency of singlet oxygen generation in the discharge. For that, the original double discharge plasma-chemical reactor has been developed. In this crossed discharge, short high voltage DBD pulses produce ionization while a comparatively low electric field supports the electric current between ionizing pulses. This allows adjusting the electric field close to the optimum for excitation of singlet oxygen molecules to increase the production yield.

### Table 2

<table>
<thead>
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<th>( G (l/min) )</th>
<th>10</th>
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<th>10</th>
<th>5</th>
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<tr>
<td>( U_{ac} (kV) )</td>
<td>5</td>
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<td>5</td>
<td>7.5</td>
<td>7.5</td>
<td>7.5</td>
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<tr>
<td>( U_{dc} (kV) )</td>
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<td>1.2</td>
<td>1.35</td>
<td>1.2</td>
<td>1.1</td>
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<tr>
<td>( I_{dc} (mA) )</td>
<td>60</td>
<td>60</td>
<td>63</td>
<td>50</td>
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<tr>
<td>Integration time (ms)</td>
<td>500</td>
<td>500</td>
<td>500</td>
<td>200 &amp;</td>
<td>200 &amp;</td>
<td>200 &amp;</td>
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</table>

#### B. Experiments with crossed discharge reactor

As a result of carried out investigation, presence of singlet oxygen has not been registered in the self-sustained barrier discharge at atmospheric and reduced pressure. However, singlet oxygen \( \text{O}_2(b^1 \Sigma_g^+) \) state has been registered in the crossed discharge at atmospheric pressure.

Experiments with the non-self-sustained crossed discharge were performed under the following conditions: atmospheric pressure, total gas flow rate 1-10 NL/min, binary mixtures \( \text{O}_2/\text{He} \) with 0.25 to 5% \( \text{O}_2 \). First of all, literature results show the need for working at high frequency to increase the yield of excited oxygen. Thus, in all experiments reported in the present paper using the crossed discharge reactor, the pulser was operated at the pulse repetition rate of \( f=25 \text{kHz} \).

Experimental conditions are summarised in table 2 and results are plotted in fig. 11 and 12 respectively. The last set of experiments plotted in fig. 13 accounts for the independent variation of the sustainer DC power.

Output voltage (and of course current) of our pulse generator are depended of load significantly. Active load is pure resistive matched load, for our generator it is \(-200 \text{Ohms} \). "No load" means generator has only external capacitor as load, and in this almost all energy is reflected back to generator. In case of real load, as crossed barrier discharge reactor, a part of energy is dissipated into load and remaining part is reflected back. The following figures present two examples and shows precisely how output waveforms of the high voltage pulse generator will be distorted. On all oscillograms, red curve is voltage (divider 1:1000), black one is current (1V=10A), magenta one is calculated power = voltage*current.

Analysis of plasma emission spectra revealed well-defined peak of excited oxygen \( \text{O}_2(b^1 \Sigma_g^+) \) at 762 nm. In fig. 10 ((left) black circle and right) the \( \text{O}_2(b^1 \Sigma_g^+) \) emission band. This assertion is consolidated by the previous spectra simulations.

Several wavelengths corresponding to atomic transitions in argon (or helium) and oxygen were used to calibrate the plasma emission spectra. The most significant atomic lines that were also detected in addition to excited oxygen in our experimental...
conditions were the 615.7 nm (OI (5D\(^\ast\)-5P)) and 844.67 nm (OI (3P-3S\(^\ast\))) atomic oxygen lines and the 728.2 nm (He (1S-1P\(^\ast\))) helium line. These lines correspond to the deexcitation of the oxygen atom in the state \(5P\), whose creation is predicted by dissociative excitation or direct impact excitation, respectively the following ways:

\[
\text{O}_2 + e^- \rightarrow \text{O}^+ + \text{O} + e^- \text{ and } \text{O}_2 + e^- \rightarrow \text{O}^+ + e^-.
\]

One can observe from fig. 11 an increase of the emission intensity of singlet sigma oxygen by a factor 2 when passing from 0.25 to 1% \(\text{O}_2\) in the binary mixture. In parallel, there is a reduction in both atomic oxygen lines and helium line. The same tendency is observed while increasing the \(\text{O}_2\) percentage in the mixture up to 5%.

Table 3

<table>
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<th>Wavelength (nm)</th>
<th>Configuration</th>
<th>Terms</th>
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<tbody>
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<td>615.7</td>
<td>2s2p(34S*)3p</td>
<td>5P - 5D(^\ast)</td>
</tr>
<tr>
<td>844.67</td>
<td>2s2p(34S*)3s</td>
<td>3S - 3P</td>
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Table 4

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<th>Wavelength (nm)</th>
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<th>Terms</th>
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<tr>
<td>728.2</td>
<td>1s2p-1s3s</td>
<td>1P(^\ast) - 1S</td>
</tr>
</tbody>
</table>

Fig. 11. Evolution of \(\text{O}_2\left(\Sigma_g^\ast\right)\), OI (615.7 nm and 844.6 nm), He (728.2 nm) emission intensities against %\(\text{O}_2\) in \(\text{O}_2/\text{He}\) mixture.

One can observe from fig. 11 an increase of the emission intensity of singlet sigma oxygen by a factor 2 when passing from 0.25 to 1% \(\text{O}_2\) in the binary mixture. In parallel, there is a reduction in both atomic oxygen lines and helium line. The same tendency is observed while increasing the \(\text{O}_2\) percentage in the mixture up to 5%.

Fig. 12. Evolution of \(\text{O}_2\left(\Sigma_g^\ast\right)\), OI (615.7 nm and 844.6 nm), He (728.2 nm) emission band intensities against %\(\text{O}_2\) in \(\text{O}_2/\text{He}\) mixture.

Left: 500 ms Integration time; right: 200 ms integration time.

Fig. 13. Dependence of the \(\text{O}_2\left(\Sigma_g^\ast\right)\), OI (615.7 nm and 844.6 nm) and He (728.2 nm) emission band intensities on DC power.

These experiments aimed to maximise the energy input into electron impact excitation of singlet sigma oxygen. One can observe from fig. 13 that there exists an excitation threshold of \(\text{O}_2\left(\Sigma_g^\ast\right)\) at \(P_{\text{DC}}\approx30\text{W}\) (\(G_{\text{ex}}=10\).
Nl/min, O2/He=1: 99). And, thereafter, an increase of singlet sigma oxygen concentration as PDC (≠ IDC) increases. This tendency tends to evolve to an asymptote after IDC=90 mA.

It is also worth noting that the emission intensity of O*, at 615.7 nm, increases with the increase of PDC, which suggests a higher O2 dissociation under these conditions.

V. CONCLUSION

O2(b′Σg+) production in self and non-self-sustained discharge in oxygen/helium mixtures at atmospheric and reduced pressure is studied. Presence of singlet oxygen has not been registered in the self-sustained pulsed barrier discharge. According to literature results and our numerical simulations, the efficiency of excitation oxygen from the ground state into singlet excited O2(b′Σg+) and O2(a′Δg) states versus reduced electric field strength E/N is demonstrated to have a maximum at E/N~10 Td. And, as only non-self-sustained discharge can exist at such an electric field that is too low for simple pulsed dielectric barrier discharge, a crossed discharge reactor was used as second experimental set up. The crossed discharge consists on a high voltage pulsed discharge and a low voltage continuous discharge. The plasma has an external source of ionization (DC current in our case, but it can be an electron beam, additional ionizing pulses and so on) and the value of E/N can be controlled independently. OES analysis revealed well-defined peak of emission of O2(b′Σg+) at 762 nm which corresponds to a O2(b′Σg+) concentration in a gas mixture with an oxygen partial pressure up to 38 Torr. Measurements in the visible of the O2(b′Σg→X′Σg) emission in the post-discharge, show that concentration in O2(b′Σg) increases with the power of the continuous discharge and increases with the decrease of the molar fraction of O2 in the flow. The last set of experiments concerns the effect of the sustainer current on O2(b′Σg+) excitation efficiency. He:O2 mixtures with 1% oxygen content reveal an increase of O2(b′Σg+) concentration with the increase of supporting discharge power.

At atmospheric pressure, ozone is effectively formed, which is a strong quencher of singlet oxygen. The discharge production of O atoms, O2 and other excited species adds higher levels of complexity of kinetics in the post discharge. Thus, the outlooks will concern experimentally, the need to work at reduced pressure with the crossed discharge, and theoretically, to develop a detailed model of kinetic processes in order to study the role of these species on singlet oxygen quenching.

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