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Classification

Physics Abstracts

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Kinetics of species produced by an electron-beam controlled discharge in oxygen at atmospheric pressure (*)

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Résumé. — L'évolution dans le temps des espèces chargées, des métastables et de l'ozone est obtenue à l'aide d'un programme de calcul. Certains résultats sont comparés à des expériences antérieures.

Abstract. — Time evolution of charged species, metastable species and ozone is obtained from a computer code. Some results can be compared with previous experiments.

The progress of gas lasers and chemical synthesis reactors using electron-beam controlled discharge requires sophisticated modelling for interpreting experiments and planning subsequent technical developments. This modelling is based on three computer codes permitting the calculation of :

— electron-beam energy branching towards the various excitation, dissociation and ionization processes ;

— discharge energy branching for the same processes ;

— time evolution of involved species densities.

The results presented in this letter have been obtained by means of the last code using data provided by the first two. The species density N_i depends upon the different species considered N_j and the reaction constants A_m (reactions of two bodies j_1 and j_2) and A_n (reactions of three bodies j_1 , j_2 and j_3). The evolution of densities N_i is given by the equations system :

$$\frac{dN_i}{dt} = \sum_m A_m N_{j_1} N_{j_2} + \sum_n A_n N_{j_1} N_{j_2} N_{j_3} + v(N'_i - N_i). \quad (1)$$

The last term of equations (1) takes into account the effect of the gas flow. v is the renewal rate. It is assumed that the density N_i is uniform in the discharge chamber and that the incoming gas with

densities N'_i is immediately mixed in the whole discharge volume. The interest of the code is its efficiency : a computation time of the order of 1 min. is enough to follow density variations of several tens of decades with time constants difference of more than ten decades. These performances are achieved by temporarily setting at equilibrium, that is $dN_i/dt = 0$ in equation (1), the species having the

Table I. — Selected reactions list. Several figures numbers do not imply an especially accurate value of the physical constant.

N°	Reaction	Rate	Refs.
1	beam + O ₂ → e + ions +	1.805 s ⁻¹	[5]
2	e + 2 O ₂ → O ₂ + ions -	1.250 × 10 ⁻³¹ cm ⁶ /s	[2]
3	e + O ₂ → O + ions -	2.109 × 10 ⁻¹¹ cm ³ /s	[2]
4	e + O ₂ → e + O ₂ ¹ Δ	4.848 × 10 ⁻¹⁰ cm ³ /s	[2]
5	e + O ₂ → e + O ₂ ¹ Σ	1.146 × 10 ⁻¹⁰ cm ³ /s	[2]
6	e + O ₂ → e + 2 O	2.274 × 10 ⁻¹⁰ cm ³ /s	[2]
7	e + O ₂ → e + O + O*	5.936 × 10 ⁻¹² cm ³ /s	[2]
8	ions - + ions + → 2 O ₂	2.000 × 10 ⁻⁶ cm ³ /s	[6]
9	O + O ₂ ¹ Δ → O + O ₂	1.300 × 10 ⁻¹⁶ cm ³ /s	[7]
10	O + O ₂ ¹ Σ → O + O ₂	3.000 × 10 ⁻¹² cm ³ /s	[7]
11	O ₂ + O ₂ ¹ Δ → 2 O ₂	2.200 × 10 ⁻¹⁸ cm ³ /s	[8]
12	O ₂ + O ₂ ¹ Σ → 2 O ₂	4.000 × 10 ⁻¹⁷ cm ³ /s	[9]
13	O ₃ + O ₂ ¹ Δ → O + 2 O ₂	3.672 × 10 ⁻¹⁵ cm ³ /s	[10]
14	O ₃ + O ₂ ¹ Σ → O + 2 O ₂	1.000 × 10 ⁻¹¹ cm ³ /s	[11]
15	2 O ₂ ¹ Δ → O ₂ + O ₂ ¹ Σ	2.000 × 10 ⁻¹⁷ cm ³ /s	[12]
16	O + O ₃ → 2 O ₂	3.786 × 10 ⁻¹⁵ cm ³ /s	[13]
17	O + 2 O ₂ → O ₂ + O ₃	3.653 × 10 ⁻³⁴ cm ⁶ /s	[13]
18	2 O + O ₂ → 2 O ₂	3.000 × 10 ⁻³³ cm ⁶ /s	[14]
19	O ₂ + O ₃ → O + 2 O ₂	9.540 × 10 ⁻²⁷ cm ³ /s	[13]
20	O* + O ₂ → O + O ₂ ¹ Σ	4.000 × 10 ⁻¹¹ cm ³ /s	[15]

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fastest evolution speeds. The algorithm validity has been tested by a classical resolution with thin steps.

The species considered in an electron-beam controlled discharge in pure oxygen at atmospheric pressure are molecular oxygen in the fundamental state (O_2), the $a^1\Delta_g$ state ($O_2^1\Delta$) and the $b^1\Sigma_g^+$ state ($O_2^1\Sigma$), atomic oxygen in the fundamental state (O) and the 1D state (O^*), ozone (O_3), positive ions (ions+), negative ions (ions-) without more detailed identification and electrons (e). The retained reactions are numbered and identified in table I. Although beam effects [1] and discharge effects [2] can be coupled more correctly, ionization is the only beam effect which has to be considered for the parameters chosen in this letter.

The experimental conditions [3] corresponding to this calculation are given in table II. Some reaction rates in table I depend on various discharge and beam properties given in table II. It should be noted that thermal dissociation of ozone (reaction 19) is quite negligible at room temperature. The variations of species densities are shown in figure 1 for the third pulse which already corresponds to the periodic regime in these conditions. With drift velocities of 218 m/s for the ions [4] and 8.2×10^4 m/s for the electrons [2], the calculated current at pulse maximum is 0.13 A/cm² to be compared with the measured value 0.12 A/cm². The average ozone concentration deduced from figure 1 is 4.8×10^{-4} to be compared with the measured value 3.8×10^{-4} . As some hypothesis can be discussed, in particular the choice of the dissociation reactions and their constants [2, 3], this agreement is partly fortuitous.

The numerical results will help defining the next experiments of ozone production whose purpose is to demonstrate the possibility of ozone production in conditions of economical interest. The model is also being used for rare gas-oxygen-iodine mixtures

Table II. — Calculations parameters.

Pressure	1.15 bar
Temperature	300 K
Renewal rate	1.5 s^{-1}
Beam voltage	100 kV
Measured beam current	8 mA/cm ²
Beam pulse full width at half maximum	24 μs
Repetition rate	0.8 Hz
Electrodes gap	2 cm
Discharge voltage	20.1 kV
Electric field/gas density	$3.6 \times 10^{-16} \text{ V cm}^2$
Discharge current density	0.12 A/cm ²
Average ozone concentration	3.8×10^{-4}

Fig. 1. — Time evolution of species density.

in view of finding the working conditions of a potential laser based on excitation transfer from $O_2(^1\Delta)$ to atomic iodine.

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