PARTICLE BALANCE IN OXYGEN DC GLOW DISCHARGE

K. Mašek and L. Láska
Institute of Physics, Czechoslovak Academy of Sciences,
Na Slovance 2, 180 40 Prague, Czechoslovakia

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ABSTRACT
Computer calculations of O_2 discharge are performed to reveal an influence of elementary processes upon the mean features of the discharge plasma. Special attention is devoted to the heterogeneous reactions of the atoms and metastables on the tube walls.

Numerical analysis of dominant production and loss processes of different particles in positive column of dc O_2- glow discharge was performed. For the discharge model the six most important species of particles were chosen: electrons, O^- and O_2^+ ions, atoms and molecules in the ground and metastable ($^1\Delta_g$) states. The transport coefficients of electrons as well as the rate coefficients for electron-molecule collisions were calculated using electron energy distribution function which was obtained by solving the Boltzmann kinetic equation. A modified algebraic form of the particle balance equations was used

$$N_i \sum_j Q_{ij} Z^P_{ij} N_j = \frac{N_i}{\tau_i^{eff}} + N_i \sum_l Q_{il} Z^L_{il} N_l$$

with an effective life time τ_i^{eff}, due to radial flows of the particles of the averaged concentrations N_i and with the correction factors Q_{ij}, due to radial distribution of the particles densities / 1 /. The production and loss of particles occur via following processes (see also in / 2 /):

P1	$e + O_2^* \longrightarrow O_2 + e$	P8	$e + O_2^* \longrightarrow O_2^+ + 2e$
P2	$e + O_2^* \longrightarrow O_2^{**} + e$	P9	$e + O_2 \longrightarrow O_2^+ + 2e$
P3	$e + O_2 \longrightarrow O_2^* + e$	P10	$O^- + O \longrightarrow O_2 + e$
P4	$e + O^- \longrightarrow O + 2e$	P11	$O^- + O_2 \longrightarrow O + O_2 + e$
P5	$e + O_2^* \longrightarrow O + O^-$	P12	$O^- + O_2^* \longrightarrow O_3 + e$
P6	$e + O_2 \longrightarrow O + O^-$	P13	$O^- + O_2 \longrightarrow O_2^- + O$
P7	$e + O_2 \longrightarrow O + O$	P14	$O^- + O_2^+ \longrightarrow O_2 + O$

The effective life time in the equations for atoms and molecular metastables has the form

$$\tau_i^{eff} = \frac{\tau_i^D}{g_i}$$

The correction factors g_i involves both the effects of a finite catalytic activity of the wall surfaces and of volume destruction processes distorting the radial Bessel distribution of the particles which is commonly used as a zero approximation / 3 /. If the radial profiles are expressed by the Bessel function of zero order for nonreflecting wall surfaces due to the radial diffusion are replaced by effective first-order reactions with the rate constants

$$\frac{1}{\tau_i^{eff}} = \frac{1}{\tau_i^{B}} = \frac{\lambda_1^2}{R^2} D_i \; ; \qquad g_i = 1$$

(λ_1 is the first root of the Bessel function, D_i is the diffusion coefficient). The correction factor Q_{ij}, occuring only in the quadratic production or loss terms, is equal to $\frac{\lambda_1^2}{4} = 1.45$ in this case. Considering a reflecting wall surface

$$n_i(r) + \chi_i \frac{R}{\lambda_1^2} \frac{dn_i(r)}{dr} = 0 \qquad \text{for } r = R$$

the factors g_i are complicated functions of τ_i^B, τ_i^W ($\tau_i^W = \frac{nV}{\frac{1}{4} n\bar{v}S\, k_i} = \frac{1}{\frac{1}{2R}\bar{v}k_i}$, it represents the time necessary for the surface destruction of all particlec in the volume V neglecting the effect of diffusion; $\bar{v}$ = mean velocity, k_i - recombination or deactivation coefficients) and other factors,
recombination or deactivation coefficients) and other factors,
expressing the influence of volume destruction processes. It was shown for several examples of the radial distribution in / 1 / (summarized in Tab. 1). The values of Q_{ij} then lie within interval $\langle 1, \frac{\lambda_1^2}{4} \rangle$. The reflection coefficient χ_i in Tab. 1 is related with the deactivation coefficient of the metastables k_D on the wall by

$$\chi_i = \frac{1}{\lambda_1} \frac{1/\tau_i^B (2-k_i)}{1/\tau_i^W} = \frac{2\lambda_1 D_i}{\bar{v}R} \cdot \frac{2-k_i}{k_i}$$

The radial negative ion flow at the wall is set equal to zero / 4, 5 /, consequently, $1/\tau_i^{eff} = 0$ in the balance equation for these ions, which we think to be a more realistic condition than the finite negative ion wall life-time of earlier papers.

To reveal the influence of the above listed processes on the macroscopic parameters of the oxygen discharge we modified the respective terms in the set of equations in the way shown in Tab. II. Some results of the numerical calculations are summarized in Figs 1 - 4 as a function of the concentration parameter NR for the constant reduced discharge current I/R = 4 mA/ /cm. The most essential changes appear in the concentrations of metastables and O^- ions as well as in the ratio $\alpha = O^-/e$. The voltage-current characteristics (compared with experimental points of Sabadil / 6 /) differ from each other much less and the curves for electrons and atoms are practically identical (these refer to the curves 1 - 4 in Figs.).

Wall surface	refl. coef.	distribut.	Q	g	$\frac{1}{\tau_i^{eff}}$
1. nonreflecting	0	$J_0(r)$ Bessel	$\frac{\lambda_1}{4}$	1	$\frac{1}{\tau_i^B}$
2. reflecting (with following dominant volume processes)					
a) diffusion	χ_i	$J_0(r)+J_1(r)$ Bessel shifted	$\langle \frac{\lambda_1}{4}, 1 \rangle$	$\frac{1}{1+\frac{\chi_i \lambda_1}{2}}$	$\frac{1}{\tau_i^B} \cdot g$
b) collisions with neutr. particles	χ_i	$\rightarrow$ Bessel with increas. χ_i	$\frac{\lambda_1}{4}$	$\sim \frac{2}{\chi_i \lambda_1} \cdot \frac{1}{\sqrt{\frac{4N}{\lambda_1^2}}}$	$\rightarrow 0$ with incr. N
c) collisions with electrons	χ_i	$\sim$ const.	~ 1	$\sim \frac{2}{\lambda_1 \chi_i}$	$\frac{1}{\tau_i^B} \cdot g$

Tab. I

Number of curve in Figs	O^-		O_2^*		O		Consider. processes	Collis. freq.
	$\frac{1}{\tau_i^{eff}}$	Q_{ij}	$\frac{1}{\tau_i^{eff}}$	Q_{ij}	$\frac{1}{\tau_i^{eff}}$	Q_{ij}		
1	$\frac{1}{\tau_B}$	Q_B	$\frac{1}{\tau_W}$	1	$\frac{1}{\tau_W}$	1	P1 - P14	/2,13/
2	$\frac{1}{\tau_B}$	Q_B	$\frac{1}{\tau_B} \cdot g_M$	Q_B	$\frac{1}{\tau_B} \cdot g_A$	Q_B	P1 - P14	/2,13/
3	0	Q_B	$\frac{1}{\tau_B} \cdot g_M$	Q_B	$\frac{1}{\tau_B} \cdot g_A$	Q_B	P1 - P14	/2,13/
4	0	Q_B	$\frac{1}{\tau_B} \cdot g_M$	1	$\frac{1}{\tau_B} \cdot g_A$	1	P1 - P14	/2,13/
5	0	Q_B	$\frac{1}{\tau_B} \cdot g_M$	Q_B	$\frac{1}{\tau_B} \cdot g_A$	Q_B	P1, P3, P6 P7, P9-P12	/2,13/
6	0	Q_B	$\frac{1}{\tau_B} \cdot g_M$	Q_B	$\frac{1}{\tau_B} \cdot g_A$	Q_B	P1 - P14	recalc.
7	0	Q_B	$\frac{1}{\tau_B} \cdot g_M$	Q_B	$\frac{1}{\tau_B} \cdot g_A$	Q_B	P1 - P17	recalc.

Tab. II

A production of O^--ions is governed by the dissociative attachment (P6) within the whole considered region of parameters NR and I/R which is balanced by the detachment either with molecules O_2 in the ground state (P11) at lower values of the parameter NR or with metastables (P12 - at lower I/R) and oxygen atoms (P10 - at higher I/R) at its higher values. As for the O_2 metastables the direct excitation from the ground state O_2 is mainly offset by the term replacing the radial flow in which the more suitable from $1/(1 + \chi_i \lambda_1/2)$ of the correcting factor g_M was used as it follows from its dependence on NR in Fig. 1. The similar situation is valid for atoms, only the dissociation is replaced by the excitation. The dissociation is considered here as a result of the balance of the collisional processes summarized above. We neglected an additional "ladder-climbing" mechanism across the vibrational excitations of the ground electronic state of O_2 which was discussed by Capitelli / 7 /. On the basis of a previous analysis and comparisons with experiments / 8 / $k_D = k_R = 10^{-3}$ was chosen in our calculations. Thus omitting inessential collisional processes (P2, P4, P5, P8) in the particle balance equations we obtained the curves number 5 in Figs (marked also by crosses).

Taking into account the set of the cross sections from / 9 - 11 / and the results of the discussion of total dissociation cross section in / 12 / we corrected some of them, especially, the cross sections with electron leading to the dissociation of O_2(we replaced that one of 4,5 eV with those, starting from 5,1 eV, 7,2 eV and 9,4 eV energies) and recalculated the electron distribution function. We again obtained a reasonable agreement of all electron transport, attachment and ionization coefficients with experiments. When the collisional frequencies of the present calculations were put in the balance equations the curves number 6 in Figs were obtained. Moreover the collisional frequencies for other additional processes, which can be considered in particle balance equations (curve 7), including those with O_3 molecules

P15	$e + O \longrightarrow O^+ + 2e$	P18	$e + O_3 \longrightarrow O^- + O_2$
P16	$e + O_2 \longrightarrow O^+ + O^-$	P19	$e + O_3 \longrightarrow O + O_2$
P17	$e + O_2 \longrightarrow O^+ + O$	P20	$e + O_3 \longrightarrow O_3^+ + 2e$

were calculated.

In conclusion, even if the precision of the collisional cross sections is limited the model of O_2 discharge should include a correct effective life time due to the heterogeneous reactions of the atoms and metastables on the wall. On the other hand, a more precise model starting from the differential form of the balance equations which would be significantly complicated by the radial drift motion of the electrons and O^- ions in a self-consistent electric field, would not bring very likely an essential improvements. However, it is expected that an augmentation of this model by the O_3 molecules in connection with volume losses of atoms could influence the results in the region of higher values of NR.

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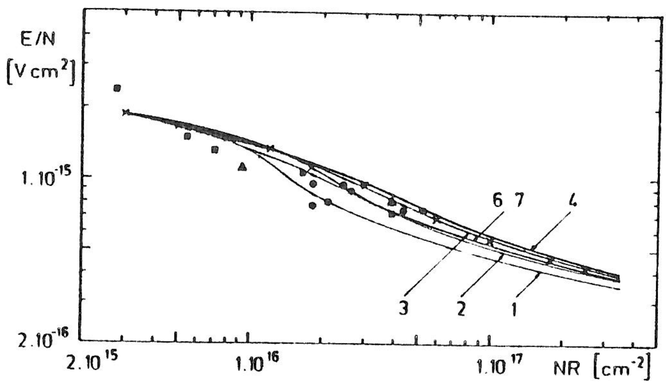

Fig. 1

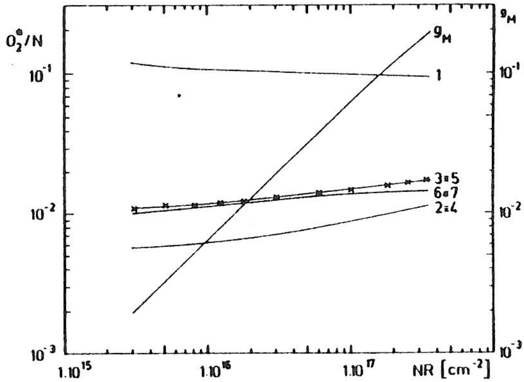

Fig. 2

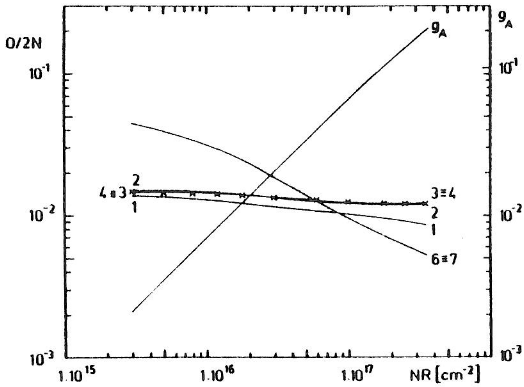

Fig. 3

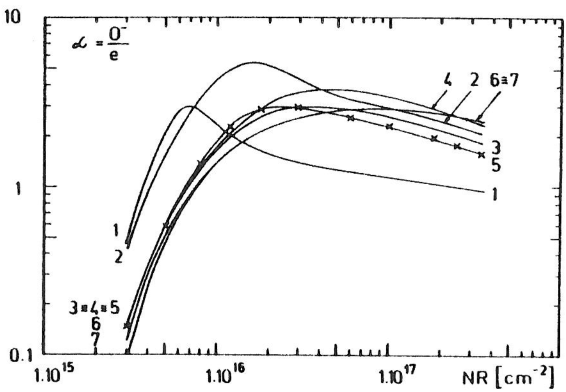

Fig. 4