

**NUMERICAL MODEL OF A GASEOUS INDUCTIVE DISCHARGE IN OXYGEN,  
TAKING INTO ACCOUNT THE COMPLETE SCHEME OF THE VIBRATIONAL  
KINETICS OF O<sub>2</sub> MOLECULES**

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**ABSTRACT**

In this work, the two-dimensional model of inductively coupled plasma discharge in oxygen was developed. The model includes hydrodynamic equations in drift-diffusion approximation for describing the kinetics of charged (electrons and ions) and neutral plasma particles, Maxwell's equations for electromagnetic fields, and equations for the temperature and neutral gas flow as well as a detailed scheme of plasma-chemical reactions. The model was tested against theoretical and experimental data in a simple cylindrical chamber. The electron density and temperature distributions were obtained over a wide range of powers deposited in the plasma (100–500 W) and compared with literature data. Also, the complete scheme of the vibrational kinetics of O<sub>2</sub> molecules was included in the model. The vibrational distribution function was calculated in low-pressure inductive discharge as well as spatial distributions of plasma parameters (density and temperature of electrons, excited components, neutral gas temperature, etc.) and flows of charged and active neutral particles at the reactor walls.

**KEYWORDS**

Low-temperature plasma; inductive discharge; oxygen; plasma modelling.

**ЧИСЛЕННАЯ МОДЕЛЬ ГАЗОВОГО ИНДУКЦИОННОГО РАЗРЯДА  
В КИСЛОРОДЕ С УЧЕТОМ ПОЛНОЙ СХЕМЫ КОЛЕБАТЕЛЬНОЙ  
КИНЕТИКИ МОЛЕКУЛ O<sub>2</sub>**

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## АННОТАЦИЯ

В данной работе разработана двумерная модель индукционного плазменного разряда в кислороде. Модель включает в себя гидродинамические уравнения в диффузионно-дрейфовом приближении для описания кинетики заряженных (электронов и ионов) и нейтральных частиц в плазме, уравнения Максвелла для вычисления электромагнитных полей, уравнения для расчета температуры и протока нейтрального газа, а также детальную схему плазмохимических реакций. Модель была протестирована на теоретических и экспериментальных данных в простой цилиндрической камере. Получены распределения плотности электронов и температуры в широком диапазоне вкладываемых в плазму мощностей (100–500 Вт) и сопоставлены с литературными данными. Также в модель была включена полная схема колебательной кинетики молекул  $O_2$ . Рассчитаны колебательная функция распределения в индукционном разряде низкого давления, а также пространственные распределения параметров плазмы (плотность и температура электронов, возбужденных компонентов, температура нейтрального газа и др.) и потоков заряженных и активных нейтральных частиц на стенки реактора.

## КЛЮЧЕВЫЕ СЛОВА

Низкотемпературная плазма; индукционный разряд; кислород; моделирование плазмы.

### Introduction

Non-equilibrium low-temperature oxygen plasma has a wide range of applications. The use of such plasma has literally revolutionised many industrial processes such as plasma etching, surface treatment, plasma sterilisation and medicine. The study of vibrational excitation and relaxation in oxygen plasma is a poorly studied area due to the complexity of experimental methods for detecting the vibrational distribution function in oxygen. New experimental data on the distribution of vibrational excitation in stationary and nonstationary discharges in oxygen was recently obtained [1], and this indicates the need to develop more accurate models to describe the relaxation kinetics of vibrational excitation under conditions of significant gas dissociation and to adapt these processes into complete self-consistent models.

The vibrational kinetics of oxygen molecules is usually not treated in global models of plasma discharges. Usually vibration-to-translation (V-T) and vibration-to-vibration (V-V) energy exchanges were taken into account and the corresponding rate coefficients were calculated, while e-V processes were considered by including only the first four vibrational levels. At that time, it was concluded that the vibrational distribution

function (VDF) was not significantly populated and it was therefore disregarded in subsequent publications. However, this picture has been challenged by recent experimental findings on a low-pressure pure oxygen discharge excited by an inductively coupled plasma (ICP) source in an industrial scale plasma etch reactor [2, 3] and recent calculations of electron-impact vibrational excitation cross-sections [4, 5]. Thus, further research in this area is necessary and self-consistent models including vibrational kinetics of oxygen molecules under the conditions of various inductively coupled discharge plasma chambers are required.

### 1. Description of the model

The two-dimensional model used in this work has been described in detail elsewhere [6, 7]. Next, we briefly describe the main points of this model without writing equations for brevity. The plasma density, the electron and ion fluxes onto the electrode, and the electron temperature were found by solving a set of following equations.

1) The continuity equations for the particle densities (electrons, ions, and neutrals).

2) The conservation equation for the particle momentum in drift-diffusion approximation both for ions and electrons.

3) The energy conservation law for electrons, taking into account Maxwellian energy distribution. For the ions, energy conservation law is not solved. In accordance with [8], it is assumed that the ion temperature was constant (on the order of 0.1–0.5 eV) throughout the chamber.

4) The distributions of the electromagnetic fields are described by Maxwell's equations, which are solved in the two-dimensional axisymmetric geometry.

5) To correctly describe the gas temperature distribution, the heat balance equation for neutral gas is solved. Accounting for gas heating is especially important when studying processes in mixtures containing molecular gases [9].

6) In order to take into account the gas flow, the Navier-Stokes equations are included in the model assuming the absence of turbulence and volumetric forces.

7) The constants of electron reactions are calculated using the energy integral of the product of the reaction cross-section, the particle rate, and the energy distribution function. In the present study, the EEDF is assumed to be Maxwellian. The experimental results [10–12] prove the validity of this assumption for O<sub>2</sub> plasma in the pressure range under study (10–20 mTorr). The electron impact and heavy particle reactions with their rate coefficients are listed in Table 1.

**Table 1.** List of electron impact and heavy particle reactions

**Таблица 1.** Список электронных и химических реакций

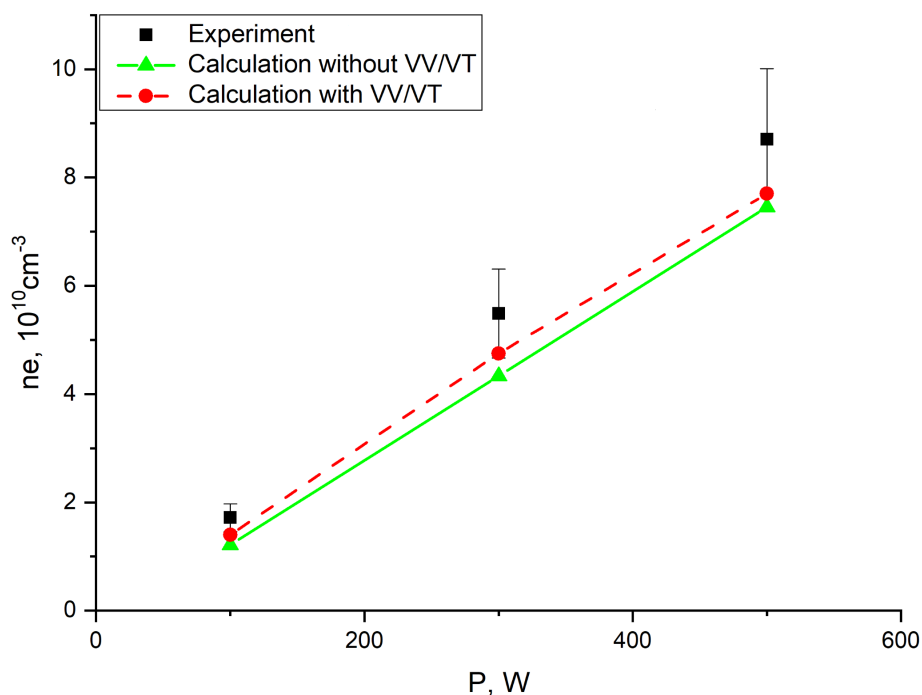
№	Electron impact reactions / Электронные реакции		
	Reaction / Реакция	Energy threshold (eV) / Энергетический порог (эВ)	Reference / Ссылка
1	$e + O_2 \Rightarrow e + O_2$	Упругое / Elastic	[13]
2	$e + O_2 \Rightarrow O^- + O$	–	[13]
3	$e + O_2 \Rightarrow e + O_2(\text{rot})$	0.02	[13]
4	$e + O_2 \Leftrightarrow e + O_2(\text{ald})$	0.977	[13]
5	$e + O_2 \Leftrightarrow e + O_2(\text{b1s})$	1.627	[13]
6	$e + O_2 \Leftrightarrow e + O_2(\text{res})$	4.5	[13]
7	$e + O_2 \Rightarrow e + O + O$	6	[13]
8	$e + O_2 \Rightarrow e + O + O(1d)$	8.4	[13]
9	$e + O_2 \Rightarrow e + O + O(1s)$	9.97	[13]
10	$e + O_2 \Rightarrow 2e + O_2^+$	12.06	[13]
11	$e + O_2(\text{ald}) \Rightarrow e + O_2(\text{b1s})$	0.5	[14]
12	$e + O_2(\text{ald}) \Rightarrow e + 2O$	5	[13]
13	$e + O_2(\text{ald}) \Rightarrow e + O + O(1d)$	7.59	[13]
14	$e + O_2(\text{b1s}) \Rightarrow e + 2O$	4.08	[13]
15	$e + O_2(\text{b1s}) \Rightarrow e + O + O(1d)$	6.94	[13]
16	$e + O_2(\text{ald}) \Rightarrow O + O^-$	–	[15]
17	$e + O_2(\text{b1s}) \Rightarrow O + O^-$	–	[15]
18	$e + O_2(\text{res}) \Rightarrow O + O^-$	–	[13]
19	$e + O(1d) \Rightarrow 2e + O^+$	11.65	[16]
20	$e + O_2(\text{ald}) \Rightarrow 2e + O_2^+$	11.08	[17]
21	$e + O_2(\text{b1s}) \Rightarrow 2e + O_2^+$	10.43	[17]
22	$e + O_2 \Rightarrow 2e + O + O^+$	21	[18]
23	$e + O \Rightarrow e + O$	Упругое / Elastic	[16]
24	$e + O \Leftrightarrow e + O(1d)$	1.968	[16]

25	$e + O \rightleftharpoons e + O(1s)$	4.192	[16]
26	$e + O \Rightarrow 2e + O^+$	13.618	[16]
27	$e + O^- \Rightarrow 2e + O$	5.5	[17]
Heavy particles reactions / Химические реакции			
	Reaction / Реакция	Rate coefficient (m <sup>3</sup> /s or m <sup>6</sup> /s) / Скорость (м <sup>3</sup> /с или м <sup>6</sup> /с)	Reference / ссылка
28	$e + O_2 + O_2 \Rightarrow O_2^- + O_2$	$1.4 \times 10^{-41} (300/T_e) e^{-600/T_e}$	[20]
29	$e + O_2^+ \Rightarrow O + O$	$2 \times 10^{-13} (300/T_e)$	[20]
30	$e + O_2^+ \Rightarrow O + O(1d)$	$1.95 \times 10^{-13} (300/T_e)^{0.7}$	[21]
32	$O_2(a1d) + O_2 \Rightarrow 2O_2$	$2.2 \times 10^{-24} (T_{gas}/300)^{0.8}$	[20]
33	$O_2(a1d) + O \Rightarrow O_2 + O$	$7 \times 10^{-22}$	[20]
34	$O_2(b1s) + O_2 \Rightarrow 2O_2$	$7.4 \times 10^{-23} T_{gas}^{0.5} e^{-1104/T_{gas}}$	[22]
35	$O_2(b1s) + O \Rightarrow O_2 + O$	$8 \times 10^{-20}$	[23]
36	$O_2(res) + O_2 \Rightarrow 2O_2$	$2 \times 10^{-19}$	[24]
37	$O(1d) + O_2 \Rightarrow O + O_2$	$6.4 \times 10^{-18} e^{67/T_{gas}}$	[25]
38	$O(1d) + O \Rightarrow 2O$	$8 \times 10^{-18}$	[26]
39	$O(1s) + O_2 \Rightarrow O + O_2$	$4.3 \times 10^{-18} e^{-850/T_{gas}}$	[20]
40	$O(1s) + O \Rightarrow 2O$	$5 \times 10^{-17} e^{-300/T_{gas}}$	[23]
41	$O + O + O_2 \Rightarrow 2O_2$	$3.34 \times 10^{-42} T_{gas}^{-1} e^{-170/T_{gas}}$	[21]
42	$O + O + O \Rightarrow O + O_2$	$3.6 \times 10^{-44} T_{gas}^{-0.63}$	[26]
43	$O^+ + O_2 \Rightarrow O + O_2^+$	$2 \times 10^{-17} (T_{gas}/300)^{-0.5}$	[26]
44	$O^- + O \Rightarrow O_2 + e$	$1.9 \times 10^{-16}$	[27]
45	$O^- + O_2 \Rightarrow O + O_2^-$	$2.5 \times 10^{-20}$	[28]
46	$O^- + O^+ \Rightarrow 2O$	$2.7 \times 10^{-13}$	[29]
47	$O^- + O_2^+ \Rightarrow 3O$	$2.6 \times 10^{-14} (T_{ion}/300)^{-0.44}$	[24]
48	$O_2^- + O_2^+ \Rightarrow 2O_2$	$4 \times 10^{-13} (T_{ion}/300)^{-0.5}$	[24]
49	$O_2^- + O^+ \Rightarrow O_2 + O$	$4 \times 10^{-13} (T_{ion}/300)^{-0.5}$	[24]
50	$O_2^+ + O^- \Rightarrow O_2 + O$	$9.6 \times 10^{-14} (T_{ion}/300)^{-0.5}$	[21]
51	$O_2(a1d) + O^- \Rightarrow O_2 + O + e$	$1.5 \times 10^{-16}$	[21]
52	$O_2(b1s) + O^- \Rightarrow O_2 + O + e$	$6.9 \times 10^{-16}$	[21]
53	$O(1s) + O \Rightarrow O(1d) + O$	$5 \times 10^{-17} e^{-300/T_{gas}}$	[23]
54	$O(1d) + O_2 \Rightarrow O_2(b1s) + O$	$2.56 \times 10^{-17} e^{-67/T_{gas}}$	[30]
55	$O(1d) + O_2 \Rightarrow O_2(a1d) + O$	$1 \times 10^{-18}$	[21]
56	$O(1s) + O_2(a1d) \Rightarrow O_2(b1s) + O(1d)$	$2.9 \times 10^{-17}$	[26]
57	$O(1s) + O_2(a1d) \Rightarrow O_2(res) + O$	$1.1 \times 10^{-16}$	[26]
58	$O(1s) + O_2(a1d) \Rightarrow 3O$	$3.2 \times 10^{-17}$	[26]
Reactions of vibrationally excited molecules O <sub>2</sub> / Реакции колебательно-возбужденных молекул O <sub>2</sub>			
59	$e + O_2(v) \rightleftharpoons e + O_2(w)$	Vibrational transition threshold (eV) / Порог колебательного перехода (эВ)	[4]
60	$O_2(v) + O \rightleftharpoons O_2(w) + O, (\Delta v = v - w = 1 - 30)$	$k_{v,w}^{(60)}$	[31]
61	$O_2(v) + O_2 \rightleftharpoons O_2(v-1) + O_2$	$k_{v,v-1}^{(61)}$	[32, 33]
62	$O_2(v) + O_2(w-1) \rightleftharpoons O_2(v-1) + O_2(w)$	$k_{v,v-1,w,w-1}^{(62)}$	[33]

The complete scheme of the vibrational kinetics of  $O_2$  molecules includes neutral particles such as unexcited oxygen molecule and atom ( $O_2$ ,  $O$ ), electronic excitations of oxygen molecule ( $O_2(a1b)$ ,  $O_2(b1s)$ ,  $O_2(res)$ ) and atom ( $O(1s)$ ,  $O(1d)$ ), vibrational excitations of oxygen molecule ( $O_2(v=1-34)$ ) and charged particles such as positive and negative ions of oxygen molecule and atom ( $O_2^+$ ,  $O_2^-$ ,  $O^+$ ,  $O^-$ ). Here  $O_2(res)$  is the sum of higher level electron excitations of the  $O_2$  molecule. Also,  $O_2(v=34)$  includes higher level vibrational excitations of the  $O_2$  molecule. It should be noted that vibrational cross-sections were summarised and normalised to the total vibrational cross-section (available, for example, at [34]) to achieve self-consistency of the resulting cross-sections set.

## 2. Verification of the model

The model was tested against the available theoretical and experimental data on discharges in oxygen from the work [35]. The calculations were performed in a simple two-dimensional axial symmetric geometry of a cylindrical reactor with a flat coil on the top of the chamber. The cylindrical discharge chamber was 16.2 cm in radius and 10.4 cm in height. The antenna was a flat four-turn coil separated from the discharge by a 1.5 cm thick quartz glass. For more details, please refer to [35]. The inductive discharge was simulated under the following conditions: the frequency was 13.56 MHz, the gas pressure was 10 mTorr, the power applied to the coils was from 100 to 500 W, and the gas flow was 33.5 standard cubic cm per minute (sccm). Part of the calculation results is shown in Fig. 1.



**Fig. 1.** Electron concentration depending on the discharge power, calculated according to a simplified kinetic scheme and taking into account vibrational kinetics, in comparison with the experiment

**Рис. 1.** Концентрация электронов в зависимости от мощности разряда, рассчитанная по упрощенной кинетической схеме и с учетом колебательной кинетики, в сравнении с экспериментом

This figure shows the electron concentration measured experimentally in work [35] using the Langmuir probe (black squares) and calculated using the model from this work (lines). All data presented in this work were obtained in the centre of the chamber, according to the position of experimental measurements. As can be seen from the figure, the model without taking into account the vibrational kinetics gives rather good agreement both qualitatively and quantitatively with the experiment within the error. Considering the complete vibrational kinetic scheme will be discussed in the next section.

### 3. Inclusion of vibrational kinetics in the model

After successful verification of the model without taking into account vibrational kinetics, the next goal of this work was to integrate the full set of reactions associated with VV/VT (Vibration-Vibration and Vibration-Translation) transfer. To do this, in addition to the reactions of VV/VT transfer, it is necessary to add to the kinetic scheme the reactions of vibrational excitation by electron impact. In Table 1 these reactions numbered 59–61. In this work 34 vibrational levels of the  $O_2$  molecule were added, based on previous calculations performed by our group. As mentioned above,

all cross sections of vibrational levels [4] were summed and then normalised to the total cross section of vibrational excitation. Then, each section was individually scaled in accordance with the obtained coefficient. In total about 2000 reactions are included in the model with full vibrational kinetics.

After these changes in the model, it was necessary to verify that the plasma characteristics did not vary. As can be seen from Fig. 1, the electron concentration remained practically unchanged (red dotted line), the change in values does not exceed 10%. Thus, the addition of a full set of vibrational kinetics did not affect the description of the main plasma characteristics by the model.

The next task of this work was to obtain the vibrational distribution function (VDF). To do this, a series of calculations was made in the conditions and geometry of the reactor from work [1]. Briefly, a pure oxygen plasma in a cylindrical chamber of 27.5 cm in radius and 10 cm in height is excited by 13.56 MHz power (up to 500 W) through a four-turn planar spiral antenna placed on a top window made of  $Al_2O_3$ . All other surfaces are hard anodized aluminium. The measurements were carried out at pressures of 10, 20 mTorr at the centre of the reactor. The results from [1] and calculations from this work are presented in Fig. 2.

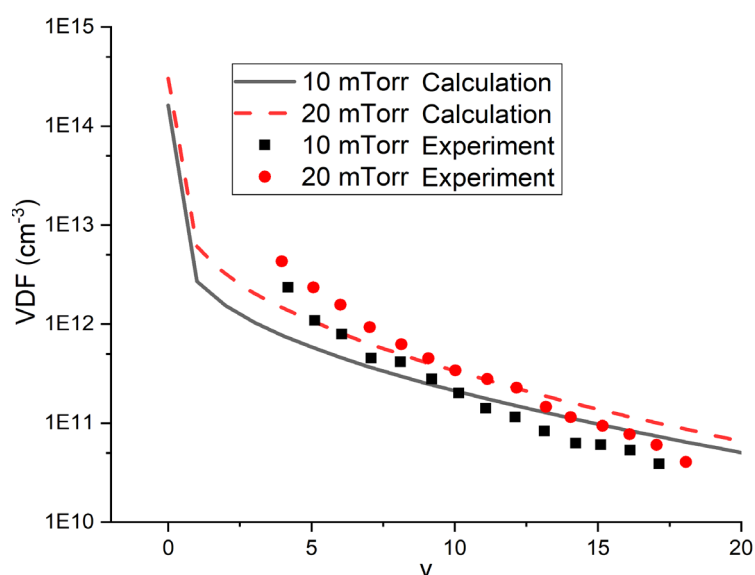


Fig. 2. Vibrational distribution function for two pressures. Experiment [1] and calculation

Рис. 2. Колебательная функция распределения для двух давлений. Эксперимент [1] и расчет

Comparison gives good agreement between experiment and calculations. A flat plateau was obtained according to experimental observations, which is impossible without inclusion of VV/VT reactions in the calculation. Its origin lies in electron impact e-V collisions and not in V-V up-pumping, in contrast to what happens in all other molecular gases known to date [1]. The relaxation of vibrational quanta is mainly due to V-T energy-transfer collisions with O atoms [1]. So the 59–62 reactions are crucial for the shape of VDF in oxygen ICP plasma. Without these processes we could not obtain the experimental shape of VDF.

### Conclusions

In this work, a two-dimensional hydrodynamic model of a plasma discharge in oxygen in two cylindrical reactors was constructed and tested. The calculations were carried out at various powers in the coils and pressures. The model showed good agreement with the experimental data. The plasma characteristics presented in this work on the example of the electron density are well described by the model both with and without detailed vibrational kinetics. However, it is impossible to achieve an experimental VDF profile without taking into account the VV/VT processes; therefore, the calculations were carried out taking them into account and the VDF were obtained for two pressures. In the future, the model can be used to obtain fluxes of charged and active neutral particles on the surfaces, as well as for plasma diagnostics, when it is impossible to carry out experimental measurements.

### Acknowledgments / Благодарности

*The work was supported by the Russian Science Foundation, project no. 23-22-00196.*

*Исследование выполнено за счет гранта Российского Научного Фонда (РНФ №23-22-00196).*

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