

# ACTINOMETRIC STUDY OF OXYGEN DISSOCIATION IN ICP SOURCE

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Results of actinometric study of oxygen dissociation in ICP source are presented. For atomic oxygen the emission lines of 844.6 nm was chosen having low impact of dissociative excitation. Actinometer gas in the experiments was argon, with emission line of 750.4 nm. Measurements of the degree of oxygen dissociation were conducted depending on the RF power supplied to the ICP source as well as on the oxygen gas pressure. The dissociation degree varies from few percent to 40 % depending on gas pressure.

PACS: 52.70.Kz

## INTRODUCTION

Weakly ionized highly dissociated plasmas are nowadays widely used for processing of different materials. Oxygen plasma is often applied in different technologies such as discharge cleaning (degreasing), plasma activation of organic materials, selective plasma etching of composites with a polymer matrix, plasma sterilization of delicate components for medical application and plasma-synthesis of nano structured materials [1, 2].

It is known that the plasma activation of the reactive gas during reactive magnetron sputtering has a dramatic effect on improving stoichiometry of synthesized oxide coatings [3], however, the physical mechanism of this activation has not yet been studied sufficiently for reasonable selection of the plasma source parameters to activate the reactive gas effectively.

One of the possible explanations of the activation effect is that the neutral oxygen atoms play the dominant role in surface reactions. Several methods for measuring the O atom density are known. Mass spectroscopy is a suitable method as long as plasma is created at very low pressure. However, the neutral oxygen atoms tend to recombine on the way from the discharge to the mass spectrometer so the measured value does not reflect the value in plasma. In contrast, optical emission spectroscopy is alternative technique providing information just from the plasma volume.

In order to find the contribution of molecular gas dissociation process to the mentioned plasma activation effect spectrometric investigation of optical emission from ICP source with Ar-O<sub>2</sub> filling was performed. This paper presents the results of experimental measurements of atomic oxygen density and the degree of dissociation of the molecular filling gas using actinometry technique.

## 1. EXPERIMENTAL SETUP

Schematic diagram of the experimental setup used in our investigation is shown in Fig. 1. The cylindrical discharge vessel has a radius  $R = 7$  cm and height  $L = 6$  cm. The sidewall of the vessel is made of metal. The glass top cover and the inductive coil are cooled by

air flow created by a fan. The vessel is evacuated by a turbo molecular pump down to a base pressure of about  $10^{-6}$  Torr. The experiments are performed in the work gas pressure range 0.5...100 mTorr.

The RF field is induced by a three-turn spiral copper coil with variable radius. RF power in the range 50...800 W at 13.56 MHz is coupled to the coil via a matchbox.

The measurements of the main plasma parameters have been led by means of the Langmuir cylindrical probe of diameter  $D_p = 0,1$  mm and length of  $L_p = 5$  mm. The probe data processing was done using the "PLASMAMETER" device. The sidewall flat probe of 1 cm<sup>2</sup> was mounted on the chamber side wall (see Fig. 1). The ion saturation regime was used.

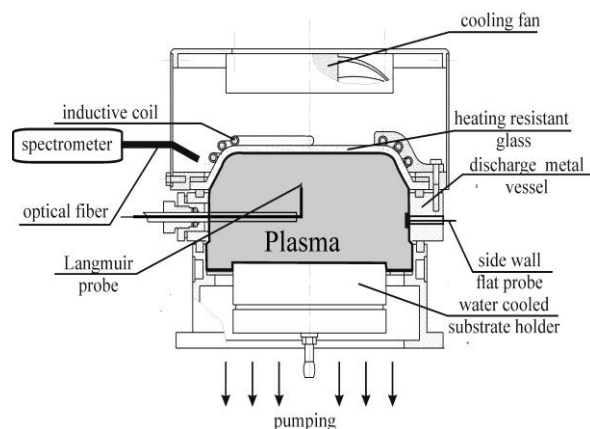


Fig. 1. Schematic diagram of the experimental set-up

In order to find the density of oxygen atoms spectrometric investigation of optical emission from ICP source with Ar-O<sub>2</sub> filling was performed. In all the experiments the Argon partial pressure was 20 % of the total gas pressure. For measurement of optical emission spectra a spectrometer with spectral resolution of about 1 nm in wavelength range of 400...950 nm was used.

## 2. MEASUREMENT TECHNIQUE

In order to determine the concentration of ground state oxygen atoms, we have used the classical technique which consists of comparing the emission of the O(<sup>3</sup>P-<sup>3</sup>S) 844 nm transition to the emission of the Ar(2p<sub>1</sub>-1s<sub>2</sub>)

750 nm transition. The energy levels taking part in the mentioned lines emission are presented in the Fig. 2.

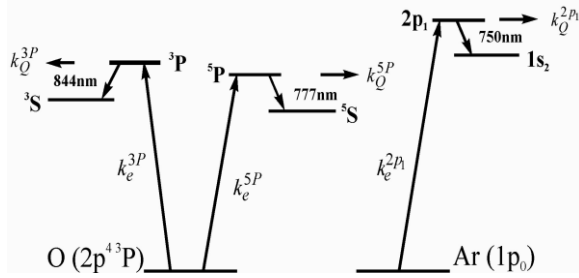
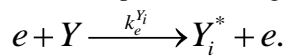
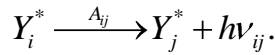


Fig. 2. The emission of the  $O(^3P-^3S)$  844 nm,  $O(^5P-^5S)$  777 nm and the emission of the  $Ar(2p_1-1s_2)$  750 nm transition

We suppose that these  $Y^*$  states are mainly populated by electron impact from the ground state  $Y$ :



The de-excitation of excited states  $Y^*$  is assumed as radiative:



The excitation coefficient  $k_e^{Y_i}$  depends on the effective electronic temperature  $T_e$  via the electron energy distribution function (EEDF):

$$k_e^{Y_i}(T_e^*) = \sqrt{\frac{2}{m_e}} \int_{E_i}^{\infty} \sigma_e^i(E) E f(E) dE,$$

with the normalization condition

$$\int_0^{\infty} \sqrt{E} f(E) dE = 1,$$

where  $E$  is the electron energy,  $\sigma_e^i(E)$  is the collision cross section with threshold energy  $E_i$  for  $i$ -level excitation,  $f(E)$  is the electron energy distribution function.

The excitation cross sections of the mentioned levels are shown in Fig. 3 for Argon and Oxygen [4] atoms.

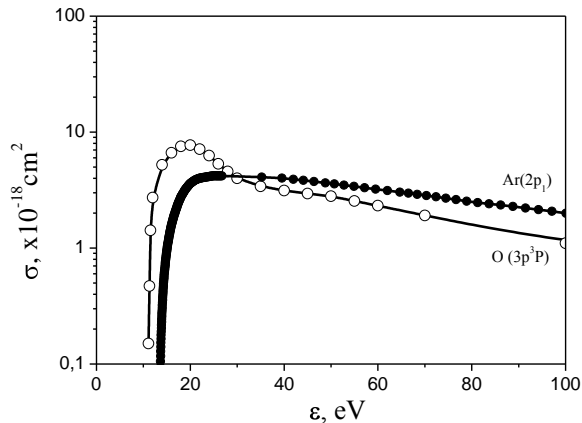


Fig. 3. Excitation cross sections of the chosen levels of Argon and Oxygen atoms [4]

The actinometry technique suppose operations with the ratios of intensities of spectral lines instead of the intensities themselves. Thus the ratio of the corresponding excitation coefficients was calculated in dependence of the electron temperature  $T_e$ . The calculation result is shown in the Fig. 4. In the experimental data interpretation we used the  $T_e$  values measured by the Langmuir probe technique.

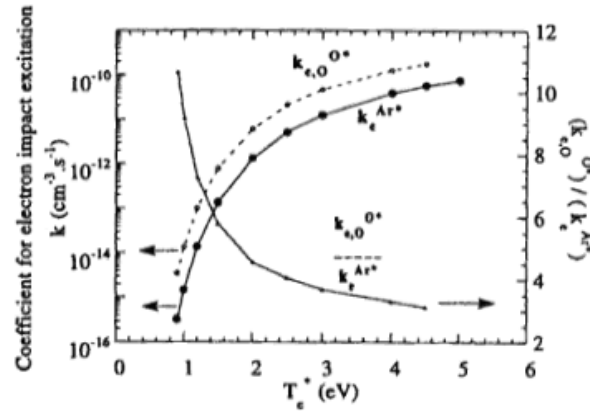


Fig. 4. Coefficient for electron impact excitation for the 750 nm argon line and 844 nm oxygen line and ration of these coefficients as function effective electron temperature

### 3. RESULTS AND DISCUSSION

The typical emission spectra at different RF power and gas pressure are shown in Figs. 5-8.

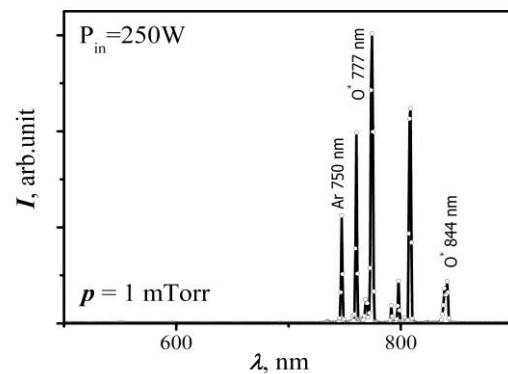


Fig. 5. Optical emission spectrum of A-O<sub>2</sub> ICP

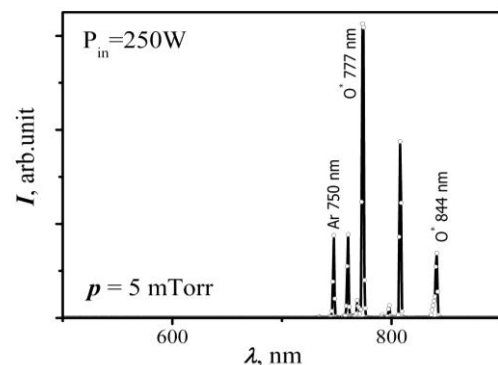


Fig. 6. Optical emission spectrum of A-O<sub>2</sub> ICP

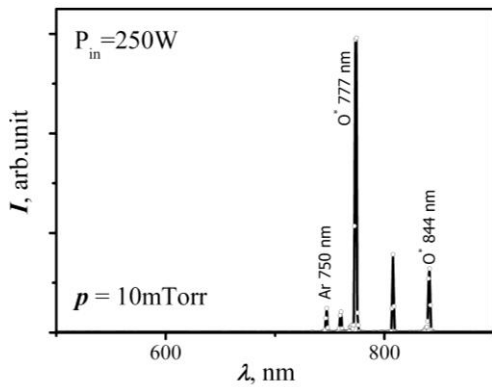


Fig. 7. Optical emission spectrum of A-O<sub>2</sub> ICP

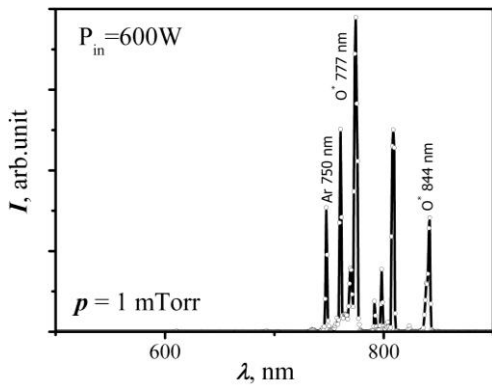


Fig. 8. Optical emission spectrum of A-O<sub>2</sub> ICP

One can see from the figures that different lines of Argon and atomic Oxygen demonstrate different dependencies on power and pressure. Using this dependencies we can define density of neutral oxygen atoms, and thus the oxygen dissociation degree. We have chosen the emission lines of atomic oxygen 844.6 nm instead of 777.3 nm having higher impact of dissociative excitation.

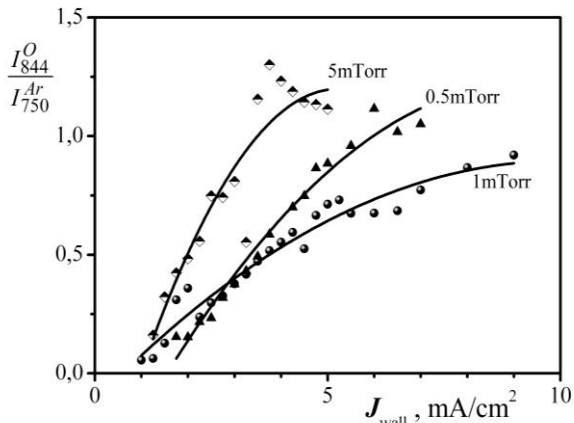


Fig. 9. The intensities ratio of O(<sup>3</sup>P-<sup>3</sup>S) 844 nm and of Ar(2p<sub>1</sub>-1s<sub>2</sub>) 750 nm, versus ion current density at the flat probe (power)

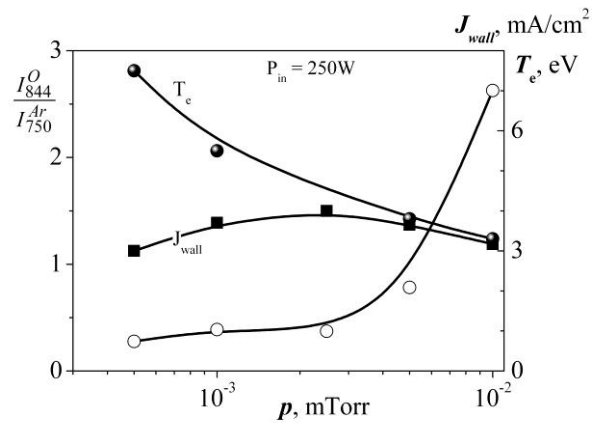


Fig. 10. Variation of intensities ratio of O(<sup>3</sup>P-<sup>3</sup>S) 844 nm and of Ar(2p<sub>1</sub>-1s<sub>2</sub>) 750 nm, electron temperature and ion current density at the flat probe from gases pressure

The main measured parameter was the line intensities ratio of O(<sup>3</sup>P-<sup>3</sup>S) 844 nm and of Ar(2p<sub>1</sub>-1s<sub>2</sub>) 750 nm. The dependencies of this parameter on the gas pressure and RF power are shown in Figs. 9, 10.

The measured ratios of the line intensities allows us to find the ratio of densities of the argon and oxygen atom sin ground state. Taking into account branching ratios for the selected radiation processes are close to 1 and using the calculated above ratio of the excitation coefficients, we can calculate the oxygen atom density, and thus the molecular Oxygen dissociation degree. The calculated oxygen dissociation degree  $\eta$  is plotted in Fig. 11 in dependence on gas pressure in the discharge chamber.

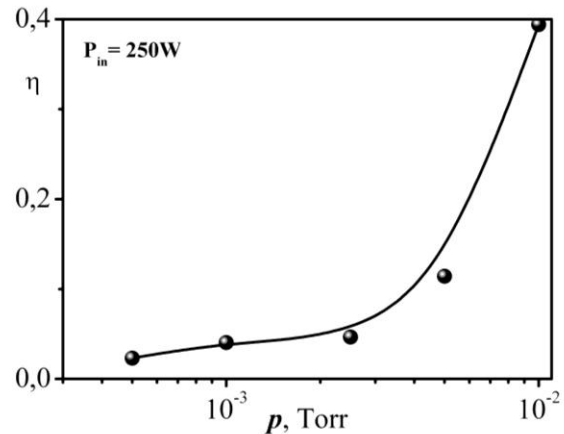


Fig. 11. Oxygen dissociation degree  $\eta$  in dependence on gas pressure in the discharge chamber

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*Article received 23.10.2014*

### **АКТИНОМЕТРИЧЕСКИЕ ИССЛЕДОВАНИЯ ДИССОЦИАЦИИ КИСЛОРОДА В ИНДУКЦИОННОМ РАЗРЯДЕ**

*А.Н. Дахов, С.В. Дудин*

Представлены результаты актинометрического изучения диссоциации кислорода в источнике плазмы на базе индукционного разряда. Для атомарного кислорода была выбрана линия излучения 844,6 нм, не подверженная существенному воздействию диссоциативного возбуждения. В качестве газа-актинометра в экспериментах использовался аргон с линией излучения 750,4 нм. Были проведены измерения степени диссоциации кислорода в зависимости от ВЧ-мощности, подводимой к источнику плазмы, а также от давления кислорода. Установлено, что степень диссоциации меняется от нескольких процентов до 40 % в зависимости от давления газа.

### **АКТИНОМЕТРИЧНІ ДОСЛІДЖЕННЯ ДИСОЦІАЦІЇ КИСНЮ В ІНДУКЦІЙНОМУ РОЗРЯДІ**

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Представлено результати актинометричного вивчення дисоціації кисню в джерелі плазми на базі індукційного розряду. Для атомарного кисню була обрана лінія випромінювання 844,6 нм, не схильна до істотного впливу дисоціативного збудження. В якості газу-актинометра в експериментах використовувався аргон з лінією випромінювання 750,4 нм. Були проведені вимірювання ступеня дисоціації кисню в залежності від ВЧ-потужності, що підводиться до джерела плазми, а також від тиску кисню. Встановлено, що ступінь дисоціації змінюється від декількох відсотків до 40 % залежно від тиску газу.