Modelling of radio-frequency breakdown in argon*

I. ILIEV, S. GOCHEVA-ILIEVA^{a*}, N. SABOTINOV^b

Department of Physics, Technical University Sofia - branch Plovdiv, 25 Tzanko Djusstabanov St, 4000 Plovdiv, Bulgaria.. ^a Faculty of Mathematics and Informatics, Plovdiv University, 24 Tsar Assen St, 4000 Plovdiv, Bulgaria ^b Georgi Nadjakov Institute of Solid State Physics, Bulgarian Academy of Sciences, 72 Tzarigradsko Chaussee Blvd., 1784 Sofia, Bulgaria

The problem of improving the agreement between the theoretically predicted and experimentally established breakdown voltage during the optimisation of the plasma technological processes is considered. New simple modified analytical breakdown criteria are obtained for low and medium-pressure RF capacitive discharge in argon at a frequency of 13.56 MHz. These criteria are based on well known criteria and experimental data and have an implicit form as a function of the parameter pd, where p is the gas pressure and d is the electrode separation. It is shown that the obtained results are in good agreement with the experiments.

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1. Introduction

Low-pressure radio-frequency (RF) capacitive discharges in gases find many applications in various plasma technological processes such as plasma etching of semiconductor materials, plasma cleaning of surfaces, deposition of oxide, nitride and other thin films, development of micro discharges for plasma displays, for chemical analysis of the materials etc. RF discharges are also used in pumping gas lasers and metal vapour lasers [1-2].

To optimize the technological processes in RF gas discharges, a detailed knowledge of the conditions for the discharge ignition is highly important. To this end specific attention is paid to measurement and simulation of the breakdown curves in direct current (DC), radio-frequency, microwave and combined electric fields. The breakdown curve is often given in the form $U_{br} = F(pd)$, where F is a function of the gas pressure p and d is the electrode separation. By varying pd we are able to design the discharge device in order to achieve better conditions for the ignition of the discharge.

The aim of this work is to use experimental data and existed criteria to obtain new simple analytical criteria, representing the breakdown voltage $U_{rf} = F(pd)$ in an RF discharge in argon.

2. Preliminary comments

Despite the fact that the breakdown in a DC discharge is due to the ion flow towards the cathode, while in RF and microwave discharges the breakdown is explained by the electron diffusion, the breakdown curves in the two cases are very similar. They are characterised by a well expressive minimum with an almost vertical left part and inclined right part. This behaviour is analogous to the Paschen curves in DC discharge.

The Townsend criterion in DC discharge is

$$1 - \gamma \left[\exp(\alpha d) - 1 \right] = 0 \tag{1}$$

where α is the coefficient of the volume electron ionization, and γ is the coefficient of the secondary emission of the electrons from the cathode [2].

The ionization of the molecules by electrons and the diffusion of electrons towards the wall of the discharge chamber play a dominant role in RF and microwave breakdown. The breakdown criterion is given by

$$\frac{\upsilon_i}{D_e} = \frac{1}{\Lambda^2} \tag{2}$$

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where v_i is the ionization frequency of the molecules from the electrons, D_e is the electron diffusion coefficient, and Λ is a diffusion length dependant on the geometry of the discharge chamber [3].

In [4], Lisovsky and Yegorenkov showed that in the presence of a weak external DC field in a RF discharge a significant increase in the breakdown voltage occurs in the right hand part of the corresponding curve (see also [3]). This is explained by electron drift towards the electrodes, which increases the loss of the charged particles. The discharge can ignite only at a higher gas voltage and pressure. The breakdown criterion in combined DC and RF fields is:

$$\frac{\upsilon_i}{D_e} = \frac{1}{\Lambda^2} - \left(\frac{E_{dc}}{2D_e/\mu_e}\right)^2 \tag{3}$$

where μ_e is the electron mobility coefficient and E_{dc} is the intensity of the DC field [4].

Criteria (1), (2) and (3) are subjects of numerous modifications [3-10]. Their purpose is to achieve a more close coincidence between the theoretical breakdown curves $U_{br} = F(pd)$ and the known experimental ones. This provides an effective tool for controlling the plasma ignition.

Argon is an important gas in laser research. It remains the focus of numerous experimental and theoretical studies [11-13]. At the same time, as discussed in [3,8], an unsatisfactory agreement between the measured and predicted breakdown voltage for this gas in RF discharge takes place.

Two basic approaches to resolve this problem are applied. The first is improving and fitting over the collision processes in order to determine the function of particle distribution or constructing different enhanced or modified analytical models with the use of molecular constants [3,5,8-10].

The second approach uses computer simulations to construct analytical models which correspond to the experimental data. Along these lines, recent papers profit from statistical methods of the PIC/MCC type (see [3,8,10] and literature quoted therein).

Bellow, we use experimental data and modify criteria (2) and (3) to obtain two simple analytical expressions, modelling the breakdown voltage U_{rf} in an RF argon discharge as a function of the parameter *pd*.

3. Construction of new modified criteria

The following theory is developed under the assumptions: 1) The RF processes are considered in a quasi-DC regime, which involves the electron energy ε , averaged over one period; 2) The coefficients D/μ and α/p , obtained for DC discharge are applicable in an RF discharge; 3) The coefficients v_d , D, D/μ and α do not change spatially and temporally.

However, recent studies [12-13] discovered that ε , v_d , D (and consequently α , which depends on ε), have complex time and spatial dependence. Despite this, we will construct analytical models that show very good agreement with the experimental data.

The RF breakdown of argon given by the criterion (2) is often represented in the form [5]:

$$\exp\left(\frac{B_0 p}{2E}\right) = A_1 p d\left(1 - \frac{E/(B_0 p)}{C_2 d/\lambda}\right) \tag{4}$$

where A_1 , B_0 and C_2 are molecular constants, p is the gas pressure, λ is the wavelength of the RF field in vacuum, $E = E_{rf} / \sqrt{2}$ is the efficiency of the RF field, and d is the electrode separation. Criterion (4) was modified in [3]. However, it is also established in [3], that (4) and its modification do not match very well the experimental breakdown results for an RF discharge in argon.

Using the approximate expression $\upsilon_i \approx \alpha v_d \approx \alpha \mu_e E$ (see [2]), from (2) we obtain $\frac{\alpha E}{D_e / \mu_e} = \frac{1}{\Lambda}$, where α is the

coefficient of the volume ionization of the gas and v_d is the electron drift velocity. Very often the coefficient of the volume ionization of the gas is given in the form [2]

$$\frac{\alpha}{p} = A \exp\left(-\frac{B}{E/p}\right).$$
 (5)

By applying different approximation formulae to the experimental data, we found that the dependence $\frac{D_e}{\mu_e} = F\left(\frac{E}{p}\right)$ is linear (with the smallest mean-squared error). For this reason, by means of the least squares method, we approximate the ratio $\frac{D_e}{\mu_e}$ in the form

$$\frac{D_e}{\mu_e} = M + N\left(\frac{E}{p}\right).$$

The diffusion length Λ for parallel plane configurations is represented by $1/\Lambda^2 = (\pi/d)^2$. Now criterion (2) becomes

$$\frac{U_{rf} p dA \exp\left(-\frac{Bpd}{U_{rf}}\right)}{M + N \frac{U_{rf}}{pd}} - \pi^2 = 0, \qquad (6)$$

where $U_{rf} = Ed$ is the breakdown voltage of the layer in an RF discharge. We obtained our first modified criterion (6) in an implicit form $F(U_{rf}, pd) = 0$ as a function of the parameter pd.

Untill now the criterion (3) was used only in the case of application of a weak external DC field. However, it is well known that the RF discharge has its own constant positive potential [2]. It may be responsible for sputtering of the electrodes in RF discharge, as is also valid in a DC discharge. Based on experimental data, in [2] the following empirical dependency between the applied interelectrode voltage U_{rf} and the voltage U_{dc} of the own DC electric field is established: $U_{dc} = U_{rf} / \pi$. Because of this we use criterion (3), without an applied external DC electric field. After some simplification (3) changes to

$$\frac{U_{rf} p dA \exp\left(-\frac{Bpd}{U_{rf}}\right)}{M + N \frac{U_{rf}}{pd}} \qquad . \tag{7}$$
$$-\frac{U_{rf}^2}{4\pi^2 \left(M + N \frac{U_{rf}}{pd}\right)^2} - \pi^2 = 0$$

For argon using the least squares method, we approximate the constants in (6)-(7) by the following values, where experimental data from [14] are used for A and B, and experimental data from [15] are used for M and N: A = 5.91189, B = 108.76559, M = 1.052994, N = 0.04039617.

4. Results and discussion

The breakdown curves $U_{rf} = F(pd)$ obtained in accordance with criteria (6) and (7) are drawn together in Fig. 1. Here, curves 1 and 2 are calculated by means of criteria (6) and (7), respectively. For comparison, in the same figure, experimental data for the breakdown voltage as a function of the parameter *pd* are also given.



Fig. 1. Comparison between the analytical criteria and
experimental data: 1 - criterion (6); 2 - criterion (7); 3 -
criterion (4);
$$\Upsilon$$
 - experimental data from [3];
 Δ - experimental data from [8]; • - experimental data
from [15].

From the figure, we can see that in practice in the entire considered interval of pd a good agreement between the suggested modified criteria and the experimental data is obtained. The two criteria are almost coincident in the left hand part of the breakdown curve in the region of low pressures. Here, the influence of the own DC electric field is very weak, and any of criteria (6) and (7) could be applied with the same accuracy. In the region of higher pressures, after the minima of the curves, the differences are more significant. The curve of criterion (7) lies in a region of higher breakdown voltages. This fact shows that the DC electric field also influences the breakdown voltage. This field causes electron loss and requires the application of a higher voltage to assure the breakdown of the discharge.

In Fig. 1 we see that at values of pd > 0.7 Torr.cm the criterion (7) describes better the experimental data given in [8,15]. In this way, our calculations show in a quality respect that at higher voltages in RF discharge the DC field must be accounted for in order to predict the breakdown curve.

We have also carried out a comparison between the criterion (4) and its modification from [3] and our criteria. The obtained results show that on the right hand part of the curves $U_{rf} = F(pd)$ criteria (6) and (7) give the same accuracy as criteria from [3].

As regards the left hand part of the breakdown curves, the criteria (6) and (7) give better accuracy than the two criteria from [3]. As an illustration, in Fig. 1 - curve 3 only represents criterion (4) by using the data from [3].

5. Conclusions

New modified criteria describing the breakdown curve in RF discharge in argon are suggested, in one of which for the first time the presence of the DC electric field in RF discharge is accounted for. It is established that in the region of the low-pressure discharges, the values of the proposed criteria (6) and (7) are very close to the experimental data, and have equal accuracies in the left hand part of the breakdown curve.

At higher pressures, for pd > 0.7 Torr.cm, it is recommended to use the criterion (7), which includes the influence of the own DC electric field in an RF discharge.

Our results are compared with well known experimental data from the literature on the breakdown curve and very good coincidence between them is established. We also carried out a comparison with other existing analytical criteria in an RF discharge. It was established that our results gave an equal approximation and furthermore were more accurate in some regions of the breakdown curve. An additional advantage of criteria (6) and (7) is their simplicity and relatively easy use in of calculations, compared to statistical models based on the PIC/MCC and other methods. The suggested criteria could be successfully applied for a fast preliminary estimate in optimizing the plasma technological processes. Such an approach in obtaining new breakdown criteria has some universality since it is fully applicable to other gasses or combinations of gasses, if sufficiently reliable experimental data are available.

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References

- M. A. Lieberman, A. J. Lichtenberg, Principles of Plasma Discharges and Materials Processing, John Wiley & Sons, New York, 2005.
- [2] Yu. P. Raizer, M. N. Shneider, N. A.Yatsenko, Radio-Frequency Capacitive Discharges, CRC Press, New York, 1995.
- [3] M. Radmilović-Radjenović, B. Radjenović, Plasma Sources Sci. Technol. 15, 1 (2006).
- [4] V. A. Lisovsky, V. D. Yegorenkov, J. Phys. D: Appl. Phys. 27, 2340 (1994).
- [5] T. Kihara, Rev. Modern Phys. 24, 45 (1952).
- [6] V. A. Lisovsky, S. D. Yakovin, J. Tekh. Fiz. 70(6), 58 (2000).

- [7] V. A. Lisovsky, J. Tekh. Fiz. 69(11), 25 (1999).
- [8] M. Radmilović –Radjenović, J. K. Lee, Phys. Plasmas, **12**, 063501 (2005).
- [9] M. Radmilović –Radjenović, J. K. Lee, Z. Lj. Petrović, Proc. 27-th Intern. Conf. Phenomena in Ionized Gases, Ed. E. M. Van Veldhuizen, Eindhoven, Netherlands, 121, 2005.
- [10] M. Radmilović-Radjenović, B. Radjenović, Plasma Sources Sci. Technol. 19, 45 (2007).
- [11] Z. Lj. Petrović et all, Plasma Sources Sci. Technol. 16, S1 (2007).
- [12] Z. Lj. Petrović, Z. M. Raspopović, S. Dujko, T. Makabe, Appl.Surf. Sci., **192**, 1 (2002).
- [13] D. Marić, M. Radmilović-Radjenović, Z. Lj. Petrović, Eur. Phys. J. D 35, 313 (2005).
- [14] V. L. Granovskii, Electrical Current in Gas, Nauka, Moscow, 1971.
- [15] V. A. Lisovskii, Pis'ma Zh. Tekh. Fiz. 24(8), 49 (1998).

^{*}Corresponding author: snegocheva@yahoo.com