<u> & ICEST 2004 ()</u>

Modeling The External Characteristic Of Cold-Plasma Reactors

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Abstract – The external or static volt-ampere characteristic describes the behavior of technological barrier discharge at diverse stages of development and application regimes. Various approaches to mathematical modeling of the external characteristic of cold plasma reactors creating plasma volumes and plasma surfaces for plasma surface modification of polymers and polymeric materials are examined.

Keywords - External characteristic, Cold-plasma reactor, Oneatmosphere barrier discharge, Plasma surface modification.

I. INTRODUCTION

Barrier discharges feature serious technological advantages determining their application to the technology of textiles and textile fibers, electronics and microelectronics, and printing industry [1].

Characteristic of all types of barrier discharges is the presence of one or two dielectric barriers separating the electrodes from the working medium.

This remains a purely external token of barrier discharges, as the dielectric barrier performs a quite essential part in discharge occurrence and burning, which is expressed by the following, [1, 2]:

 \Box the barrier with its capacitance C_{δ} performs the part of a reactance, i. e. of a capacitive ballast reactance $X_C = \omega^{-1} C_{\delta}^{-1}$, that limits the increase of the electric current in the course of discharge burning;

 \Box the barrier re-distributes the electric field intensity in the interelectrode space, loading electrically the working (gaseous or vaporous) medium, the intensity in this medium increasing ε_{δ} times with respect to the electric field intensity in the barrier, where ε_{δ} is the relative dielectric permittivity of the barrier material or it determines the critical parameters of barrier discharge: voltage of ignition U_{bd} and current of ignition I_{bd} ;

 \Box the barrier determines the voltage of burning U_b of the discharge, which remains constant in the course of its burning and does not depend on the working voltage chosen.

¹ The multiple ionization-related and chemical processes going simultaneously during burning of the barrier discharge at atmospheric pressure present considerable problems not only in controlling of discharge, but also in describing its behavior, [2].

THE TASK of the present work is to use various approaches to modeling the behavior of a low-frequency

(50 Hz) barrier discharge, which burns in the volume or on the surface of a cold-plasma generator system, under load or in the absence of a load, in air at atmospheric pressure (**760** \pm 25 Torr, **1** atm), i. e. of a *one-atmosphere barrier discharge (OABD)*.

The investigations are focused on two types of a cold-plasma reactor system:

◆ *the first one* representing two plane-parallel electrodes with a glass barrier between them, which create a plasma volume with relatively uniform distribution of the electric field between the glass barrier and one of the electrodes, i.e. a *one-atmosphere uniform barrier discharge (OAUBD)*;

◆ *the second one* representing two plane-parallel electrodes, which embrace tightly the glass barrier in such a way, that solely on that side of the barrier, which looks at the combshaped electrode, there emerges a plasma surface with strongly non-uniform distribution of the electric field, i. e. a *one-atmosphere non-uniform barrier discharge (OANUBD)*.

II. General formulation of the investigations

Our experimental investigations [2, 3, 5] performed during a continuous period of time allow to search for a new description of the behavior and control of the barrier discharge at atmospheric pressure, based on its external or static voltampere characteristic.

This characteristic expresses the relationship between the average value of current I_{gap} (AV) passing through the barrier discharge and the effective value of voltage U_{gap} (RMS) applied across discharge, Fig. 1.

Moreover, it turns out [2,3], that the external characteristic may be simulated by means of a broken-line polygon of three linear segments, each of them corresponding to one of the three development stages of the barrier discharge, Fig. 1:

♦ the stage preceding the ignition of the barrier discharge, namely the so-called free or non-operational regime;

♦ the first stage of burning, which corresponds to the formation of cold ozone- and oxygen-containing plasma;

• the second stage of burning, which corresponds to the formation of cold plasma mostly containing nitrogen oxides.

At high values of linear correlation factor r_{pc} the linear law describes very well the individual sections of the external characteristic of barrier discharge. However, the transitional regions of the characteristic remain outside the scope of this description, because there is a smooth transition between each two adjacent regions, while the polygon simulating the characteristic represents a broken line.

Do the latter two stages (or regimes) of burning of the barrier discharge really exist? The answer is positive, because the analysis of the elementary processes clearly separates from each other the two air media, in which ozone and products of its decomposition are generated, [1]:

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• one of these, a medium depleted of energy, is created in electric fields of low relative intensity E/p, favoring processes with larger cross-section of impact interaction – about 10^{-16} cm², i. e. ozone generation from molecular ions O_2^+ , where two molecules of ozone O_3 correspond to one molecular ion O_2^+ . A product of its rapid decomposition under conditions of α -impact ionization are the exited, chemically active oxigen (O_2^+) molecules (the so-called singlet oxygen);

• the other, energy-rich medium, is created in electric fields of higher relative intensity E/p and favors processes with smaller cross-section of impact interaction – about 10^{-18} cm², i. e. the generation of ozone O_3 from atomic ions O^2 and O^+ through intermediate synthesis of negative O_3^- and positive O_3^+ ozone ions.

The energy–richer medium of higher values of the relative electric-field intensity E/p and plasma temperature creates conditions not only for rapid decomposition of ozone, but also for its inhibition in reactions with negative atomic ions O^{-} , with atomic oxygen O, or with nitrogen oxides NO_x . Such a medium is characterized by rapid depletion as regards the ozone and products of its decomposition. It is enriched with nitrogen oxides, [1].

This thesis is supported by the direct measurement of the ozone obtained in ozone-air mixtures in the two regimes of burning of the barrier discharge, [4].

The mathematical model thus obtained is based on its real reasons, moreover that experiments in modifying low-energy polymeric surfaces indicate different physico-chemical relations of materials in these two cases, [4].

This comes to show that there exist physical, chemical and technological reasons that make this model very useful. Oneatmosphere barrier discharge can be simulated and controlled as a voltage-controlled current source.

At the same time *Zhiyu Chen* and *J. Reece Roth* [5] propose a new model of behavior of one-atmosphere barrier discharge based not on the linear law, but on a power one. The discharge current follows a power law, a characteristic of the voltage-current behaviour of a normal glow discharge (in vacuum). For example, the current-voltage relationship of various normal glow discharges in vacuum was $I \propto U^2$, $I \propto U^3$, and $I \propto U^9$.

Its output current follows the power law given by the following equation

$$I = 0, \text{ for } U_{gap} < U_{bd} \tag{1}$$

$$I \alpha \left(U_{gap} - U_{bd} \right)^n, \text{ for } U_{gap} \ge U_{bd}$$
(2)

where *n* is an integer that may range from 1 to 12 in different types of glow or barrier discharge plasma devices, U_{bd} - the voltage of breakdown.



Fig. 1. Operating sectors on the external characteristic of oneatmosphere barrier discharge, namely the relationship between the average value of current I_{gap} and the effective value of applied voltage U_{gap} .

OA - non-operating sector; AB – first stage of burning – a cold technological plasma containing ozone and products of its decomposition; CD - second stage of burning – a cold technological plasma containing nitrogen oxides; BC – transient area.

A power law, for which $U_{gap} < U_{bd}$ is set equal to zero, does not adequately simulate the behaviour of the oneatmosphere barrier discharge. The power law relates to the whole region of burning of barrier discharge, i. e. for $U_{gap} \ge U_{bd}$.

Furthermore, the authors also assume that the voltage of burning U_b of barrier discharge remains constant, irrespective of the value of voltage U_{gap} applied across discharge gap, and for this reason they also speak for one-atmosphere *glow* discharge plasma (*OAGDP*).

In the present work it is made an attempt to transfer the new relationship found between the instantaneous values of current and voltage onto the simulation of the experimentally obtained external characteristic or between the average value of current I_{gap} and the effective value of voltage U_{gap} . This is a possibility of obtaining a unitary controlling model for the whole region of burning of barrier discharge.

The generalized model of burning of one-atmosphere barrier discharge is created under the following conditions:

 \Box the barrier discharge, similarly to the normal glow discharge in vacuum, burns at a constant value of the voltage of burning, i. e. $U_b = const$;

□ the ignition of barrier discharge represents a threshold process occurring for certain critical parameters – the voltage of ignition U_{bd} (max) and current of ignition I_{bd} (AV), this determining the necessity that the description of discharge in the stage of burning is governed by eq. 2;

□ the barrier discharge may be described in the whole region of burning or individually in each of the two sub-regions of burning;

☐ the generalized equation of barrier discharge burning, governed by the power law, is of the following form:

$$\frac{I_{gap} - I_{bd}}{\left(U_{gap} - \frac{U_{bd}}{\sqrt{2}}\right)^n} = B$$
(3)

where I_{gap} (*AV*) is the current through the barrier discharge; U_{gap} (*RMS*) – the voltage across discharge gap; I_{bd} (*av*) and U_{bd} (*max*) – the critical ignition parameters of discharge; $n \ge 1$ – the exponent; B – a constant determining the increase of discharge current.

 \Box the generalized equation includes in itself the linear law of variation as a particular case at n = 1:

$$\frac{I_{gap} - I_{bd}}{\left(U_{gap} - \frac{U_{bd}}{\sqrt{2}}\right)} = B = tg\alpha \text{, or}$$
(4)

$$I_{gap} = B U_{gap} + A , \text{ at}$$
 (5)

$$A = I_{bd} - B \frac{U_{bd}}{\sqrt{2}}.$$
(6)

II. RESULTS AND DISCUSSION

A. General presentation of the regions of burning

The experimental investigations are conducted with a barrier representing a plate of thickness $\delta = 3$ mm, made of alkaline silicate glass of dielectric permittivity $\varepsilon = 10$, volumetric specific resistance $\rho = 10^9 \Omega m$ and $t_g \delta = 25$ (at 20 °C).

External characteristics of the type $I_{gap}(AV) = \varphi [U_{gap}(RMS)]$ are obtained experimentally for two types of plasma generator systems:

♦ the first one having a virtually uniform electric field of a one-atmosphere uniform barrier discharge – the so-called OAUBDG-system; and

♦ the second one having a non-uniform electric field of a one-atmosphere non-uniform barrier discharge – the so-called OANUBDG-system.

The *OAUBDG*-system creates a plasma volume between the glass barrier and one of the two planar metal electrodes of area $S = 651.5 \text{ cm}^2$ and shape reducing the edge effect, the thickness of the plasma region being H = 6 mm. The barrier capacitance is $C_{\delta} = 1192 \text{ pF}$, measured at industrial frequency.

The *OANUBDG*-system creates a plasma surface on one of the two flat metal electrodes of area $S = 480 \text{ cm}^2$ that embrace tightly the glass barrier. The discharge burns on the side to the electrode made in the form of a comb with width of 4 mm of its constituent elementary electrodes and a distance of 4 mmbetween each two of them. The discharge burns on the dielectric barrier itself – between the elementary electrodes. The barrier capacitance is $C_{\delta} = 536 \text{ pF}$ measured at industrial frequency.

The experimentally obtained external characteristics (for both plasma systems) permit applying both approaches to modeling in no-load regime. The linear model for the ozone-oxygen region of burning the barrier discharge has the following parameters in accordance with eq. 5, Table 1.

TABLE 1.

System	Β , μΑ/k V	Α , μΑ	$oldsymbol{U}_{bd},\mathrm{kV}$	I _{bd} , μΑ	Correla- tion Coef- ficient r _{lc} , /
OAUBDG	19.58	- 0.55	7.700	151	0.999375
OAUNBDG	349.7	- 455	3.014	599	0.998380

The linear model for the second region (that of the nitrogen oxides) of discharge burning has the following parameters in accordance with eq. 5, Table 2.

TAB	le 2
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TABLE 4.

System	B , μA/kV	Α , μΑ	U _{bd} , kV	Ι _{bd} , μΑ	Correlation Coefficient r _{lc} /
OAUBDG	26.82	- 62.5	11.92	250	0.993858
OAUNBDG	399.0	- 1145	14.00	4441	0.998640

The model generalized according to the power law for the whole region of discharge burning has the following characteristical parameters in accordance with eq. 3, Table 3. TABLE 3.

System	U _{bd} , kV	<i>I_{bd}</i> , μΑ	INTEGER n, /	Correlation Coefficient r _{lc} /
OAUBDG	7.700	151	1.5	0.99467
OAUNBDG	3.014	599	1.1	0.99934

In loading the *OAUBDG*-system almost the whole plasma volume is filled with the material to be treated; in this case this is a non-woven textile based on polyethylene terephthalate (PET) with area mass 500 g/m^2 . For this type of textile load the external characteristic changes to a significant extent – the region of burning is a single one and corresponds to the first characteristic region of burning, Table 4.

System - Model	B , μA/kV	Α , μΑ	U _{bd} , kV	I _{bd} , μΑ	Correlation Coefficient r _{lc} , /
OAUBDG- Linear model	359.37	- 372	10.85	172	0.992730
System - Model	INTEGER n, /		U _{bd} , kV	<i>I_{bd}</i> , μΑ	Correlation Coefficient r _{lc} /
OAUBDG- Power law	1.47		10.85	172	0.999195

Analyzing the models obtained indicates that both approaches enable making a description of the process by regions of burning or for the whole region of burning at a relatively high value of the linear correlation factor. This makes possible the application of both approaches for the purposes of examining or controlling the technological process.

Describing fully the process of discharge burning by means of a single function is of great practical importance, as it allows investigating very easily the influence of various parameters of the plasma generator system upon exponent n as

well as upon two parameters critical for the ignition - I_{bd} and U_{bd} , Table 3.

Moreover, this characteristic may be made linear by taking the logarithm of both sides of eq. 3:

$$lg\left(I_{gap} - I_{bd}\right) = n \, lg\left(U_{gap} - \frac{U_{bd}}{\sqrt{2}}\right) + lg \, B \,. \tag{6}$$

The linear model is suitable for making an individual description for each of the two characteristic regions of burning of the discharge. This is especially imperative in the case of realizing a technology in only one of the two technological regions of burning.

A. Using the power law model for describing the individual regions of burning of the discharge.

The values of the linear correlation factor are not always high, i. e. above 0.96, all over the investigated region, Fig. 2.



Fig. 2. There are regions in the space examined – thickness of the dielectric barrier δ and size of plasma volume *b*, of the first sub-region (or regime) of burning without load of the *OAUB*-discharge, in which linear correlation factor r_{lc} is of relatively low values, namely below 0.96.

This situation can be changed if using the generalized law for describing those individual regions of burning of the discharge, which are characterized by lower degree of linearity of the external characteristic.

As an example it is taken an *OAUBDG*-system with barrier thickness $\delta = 7 mm$ and largest size of plasma volume H = 9 mm, operating in a regime under load: treating PET non-woven textile with area mass $500 g/m^2$ and thickness 5 mm. At n = 1 the linear correlation factor is relatively low, namely 0.9898, Table 6.

Verifying linear correlation factor r_{lc} for various values of exponent *n* indicates a new, higher value of $r_{lc} = 0.9960$ at n = 1.66, Table 6.

This is another possibility of describing more precisely the two technological sub-regions of burning of the oneatmosphere barrier discharge.

III. CONCLUSION

By using both methods investigated, it is possible to model successfully the experimentally obtained external characteristic of one-atmosphere barrier discharge with industrial frequency (50 or 60 Hz) in the region of burning, either as a whole, or individually for each of its two parts.

TABLE 5.

System - Model	B , μA/kV	Α , μΑ	U _{bd} , kV	I _{bd} , μΑ	Correlation Coefficient r _{lc} , /
OAUBDG - Linear model	97.56	- 1079	12.56	146	0.9898
System - Model	Integer n, /	B , μA/kV	U _{bd} , kV	<i>I_{bd}</i> , μΑ	Correlation Coefficient r _{lc} , /
OAUBDG - Power law	1.66	13.7	12.56	146	0.9960

The linear law that relates the average value of current I_{gap} through the discharge to the effective value of voltage U_{gap} , applied across discharge gap, is suitable for describing and controlling the burning of discharge in its two technological sub-regions, while the power law is more suitable for involving the whole region of burning of the barrier discharge.

However, the power law may be applied with the same success to certain cases, where a more precise description of behavior in the two sub-regions of burning is necessary.

In both cases, starting from the models of the external characteristic obtained as described and performing the necessary calculations, it is possible to determine the technological characteristic of the barrier discharge.

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ACKNOWLEDGEMENT

This research was financially supported by Ministry of Education and Sciences (MON) - Project MU-TN-1201 / 02.