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LOW-FREQUENCY TECHNOLOGICAL DISCHARGE AT ATMOSPHERIC PRESSURE

Dilyana GOSPODINOVA, Peter DINEFF

*Technical University of Sofia, Department of Electrical Apparatus, 1000 Sofia, Bulgaria,
E-mail: dineff_pd@abv.bg; dilianang@yahoo.com

Abstract. The low-frequency electric discharge that burns at atmospheric pressure and industrial frequency (50/60 Hz) represents a good technological alternative of the *RF*- and glow discharges at low pressure (in vacuum). The absence of a labor-consuming and expensive technological vacuum installation is one of its biggest advantages, which stimulate its fast introduction into the practice of plasma-chemical technologies. The expected lower intensity of the technological process of modification of polymer surfaces cannot become an obstacle for its broader application.

Keywords: barrier discharge, external (volt-ampere) static characteristic, high-pressure low-frequency discharge, plasma-chemical surface modification, surface power density.

INTRODUCTION

The low-frequency technological electric discharge at atmospheric pressure and industrial frequency (50/60 Hz) is the more and more frequently used alternative of glow and *RF*-electrical discharges in the practice of plasma-chemical surface treatment of polymers and polymer materials. The absence of labor-consuming and expensive technological vacuum system and the use of voltage with industrial frequency are advantages, which, together with its technological effectiveness, are constantly widening the range of its practical applications.

The low-frequency electric discharge uses a capacitive ballast reactance to control the electrical regime. The discharge is realized mainly in two technological varieties: the *barrier* and *corona* electric discharges. The corona electric discharge is internally unstable and may easily develop into electric arc if there is enough power, which is undesirable from technological point of view.

Burning of a low-frequency electric discharge in air at atmospheric pressure is determined in the first place by the ionizations of oxygen and nitrogen, i. e. by the production of ozone and the chemical active products resulting from its decomposition, and of the nitrogen oxides and their active oxidation.

However, the variety of ionization and chemical processes is too much considerable and is an essential obstacle to describing and controlling the plasma-chemical modification.

THE TASK of the present work consists in demonstrating an original approach that allows describing and controlling the two technological regimes of plasma-chemical modification characteristic for the low-frequency electric discharge, namely the regime of ozone- and oxygen-containing cold plasma and that of cold plasma containing nitrogen oxides.

CHARACTERISTICS OF THE LOW-FREQUENCY ELECTRICAL DISCHARGE

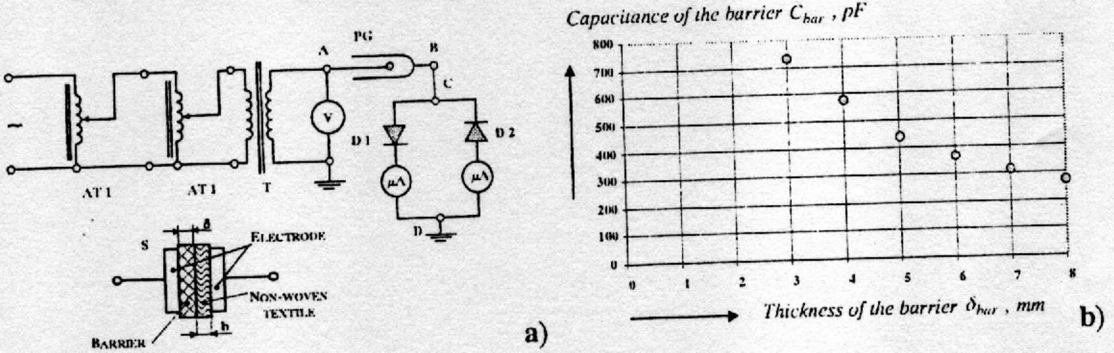
External static characteristic of discharge

In the general case, the electric energy consumed in maintaining the burning of a barrier (low-frequency) discharge is transformed into the energy of the following changes that occur concurrently in the air:

- ◆ impact dissociation - molization;
- ◆ impact ionization - recombination;
- ◆ chemical processes of oxidation and reduction.

The common trait of these three groups of processes consists in the fact that they can be perceived as processes connected with an exchange of electrons. This fact may give a serious reason for seeking a general description of the process based on the quantity of electricity carried over by the discharge (for a half-period of change), i. e. for transferring concepts connected with the fundamental laws of *Faraday* concerning the oxidation-reduction processes in electrolysis.

The electrochemical action of alternating current is effectively presented by its average value I_{av} that defines the intensity of carrying over a quantity of electricity Q through the electric discharge: $Q = I_{av} t$.



a) b)

Figure 1. Experimental setup of the investigation: **a** – electric circuit for experimental plotting of the external static characteristic of the discharge and general view of the plasma generator; **b** – experimentally determined relationship between the thickness and capacitance of glass (alkaline glass) barrier.

The experimental determination of the external static characteristic of barrier electric discharge is conducted with the help of the circuit shown in Fig. 1a. This characteristic is obtained as a relationship between the average value of electric current I_{av} and the effective value U_{eff} *of* the voltage applied to plasma generator. The variation of barrier thickness defines its capacitance and the value of the ballast reactance limiting the electric current, Fig. 1b.

When the alternating voltage varies harmonically, the external characteristic is a broken-line polygon consisting of straight segments; the first straight-line sector determines the flow of reactive current of electric conductance, and the second one characterizes the burning of barrier discharge. In the course of these investigations it has been experimentally found out there exists one more or third straight-line sector which, on its part, characterizes the burning of barrier discharge, too, Fig. 2.

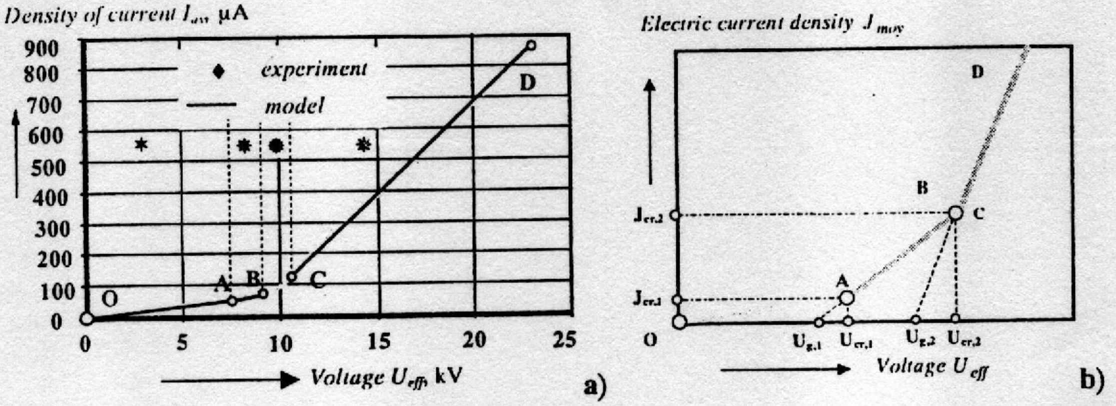


Figure 2. Operating regions of the external characteristic of electrical barrier discharge; the relationship between the average value of current I_{av} and the effective value of applied voltage U_{eff} **(a)**.

OA – non-operating sector; **AB** - first operating sector, namely cold technological plasma containing ozone and products from its decomposition; **CD** - second operating sector, namely cold technological plasma containing nitrogen oxides; **BC** - transient region. Critical electrical parameters **(b)** of the two transitions towards the first **(A-B)** and second **(C-D, point B ≡** point C) operating sectors of the external characteristic of barrier discharge.

Straight-line operating sectors *AB* and *CD* may be represented through equations of linear regression on the basis of the experimental results obtained:

$$I_{av} = B\,U_{eff} + A\,,\; B = \text{Const},\; A = \text{Const}. \tag{1}$$

This formulation has been developed on the basis of screening investigations of the no-load characteristics of plasma generator *PG*, when the air gap is empty, i. e. no test specimen is being treated. A complete investigation is realized by using an electrode system consisting of two rectangular flat-parallel electrodes with rounded ends, of a radius of 5 mm, in order to diminish the edge effect. The barrier used is made of alkaline glass with dielectric permittivity $\varepsilon = 10$, volumetric specific electrical resistance $\rho = 10^9\ \Omega.m$ (at 20 ^oC) and $tg\,\delta = 25$. Glass barrier thickness δ and size b of working air gap are perceived as parameters of the investigation.

The straight-line sectors are defined consecutively one by one, starting from the non-operating sector *OA*. New points are added successively to the set of points belonging to the sector of the external characteristic until the coefficient of linear correlation starts decreasing. This is a sign that the new point does not pertain to that straight-line sector. The well-known procedure of least squares is applied. As a result, three characteristics of the straight-line sector are determined: the rate of current increase I_{av} with voltage B, μA/kV, the intercept or free term A, μA, and the coefficient of linear correlation r.

The coefficients of linear correlation r for the whole region of investigation are shown separately for each of the two operating sectors *AB* and *CD* of the external characteristic, in Fig. 3 and Table 1. Some positions of those in Table 1 are not filled in as the third sector has not yet manifested itself fully for the discharge burning voltages used, namely up to $U_{eff} = 22$ kV. This requires especially that data be presented in tabular form.

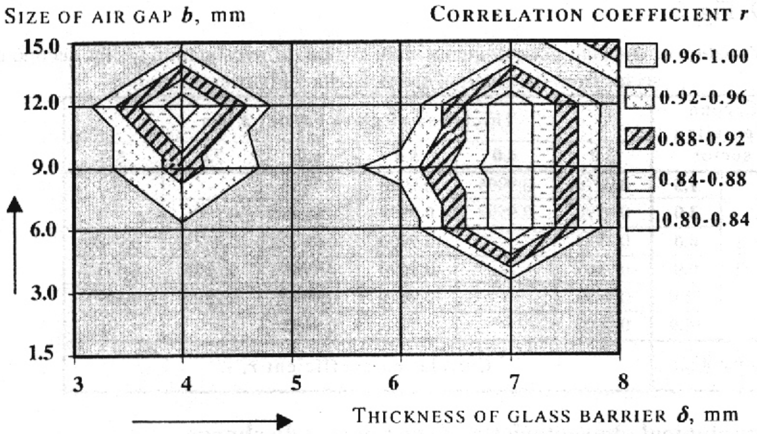


Figure 3. Variation of the coefficient of linear correlation r characterizing the connection between current I_{av} and voltage U_{eff} in first operating sector *AB* of the external characteristic, which is determined from the variation of the thickness of glass barrier δ and the size of working air gap b in the region of investigation.

The coefficient of linear correlation r assumes its highest values for small sizes of the air gaps as well as for relatively thin glass barriers, which confirms the arguments introduced so far with respect to the validity of proposed linear model for the barrier discharge behavior, Fig. 3 and Table 2.

The values of coefficient r above 0.99 demonstrate a strong, almost functional, linear relationship between the variation of current I_{av} and voltage U_{eff}. Increasing the voltage U_{eff} leads to an increase in current I_{av} or $r < 1$. The values exceeding 0.95 confirm the existence of the linear correlation in statistical sense. Values above 0.90 show a linear correlation which is still acceptable, while those below 0.90 demonstrate just a tendency related to linear correlation.

The linear relationship between the variation of current I_{av} and voltage U_{eff} can be explained solely with the constant voltage of burning U_g of the discharge and the linear variation of the voltage applied across the glass barrier reactance.

Current I_{av} and current density J_{av} cannot be technological characteristics of the glow discharge as their variation does not reflect the threshold character of the processes of dissociation and ionization, i. e. they do not take into account the voltage influence through the critical discharge parameters, Fig. 2b.

Active power P of the discharge reflects the increase in the average value of current I_{av} above its critical value I_{cr} and the discharge burning voltage U_g that remains constant during the entire operating time interval:

$$P = (I_{av} - I_{cr})U_g \,. \tag{2}$$

This form of the relationships is valid for both technological regimes of the discharge: *AB* and *CD*, Fig. 2a.

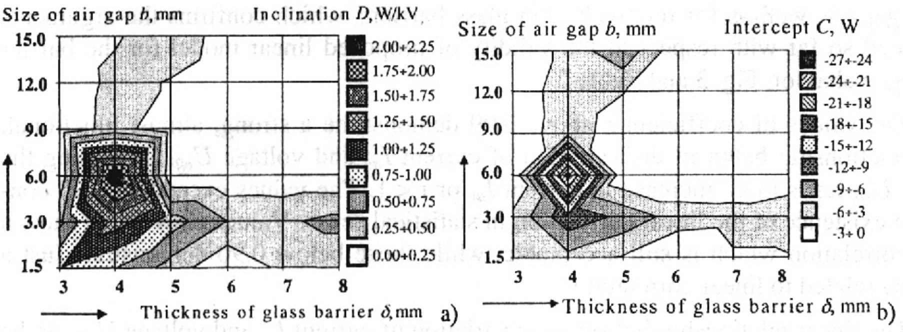
Table 1. Variation of correlation coefficients in the region of investigation for the second operating sector from the external characteristic of barrier discharge.

Second Operating sector		Thickness of glass barrier δ, mm					
		3.0	**4.0**	**5.0**	**6.0**	**7.0**	**8.0**
Size of gap *b*, mm	**1.5**	0.9976	0.9995	0.9998	0.9981	-	-
	3.0	0.9996	0.9992	0.9979	0.9998	0.9996	-
	6.0	0.9939	-	0.9902	0.9959	0.9979	0.9986
	9.0	0.9967	-	0.9690	0.9984	0.9938	0.8918
	12.0	0.9893	-	0.9679	0.9970	0.9923	0.8071
	15.0	0.9600	-	-	0.9695	-	-
$I_{av} = \varphi(U_{eff})$		Correlation coefficient *r*, -					

Technological characteristic of the barrier discharge

Furthermore, active power P, just like current I_{av}, represents a linear function of varying voltage U_{eff}:

$$P = (I_{av} - I_{cr})U_g = BU_gU_{eff} + (A - I_{cr})U_g = DU_{eff} + C \,,\; D = \text{Const},\; C = \text{Const}. \tag{2}$$



a) b)

Figure 4. Main characteristics of the first operating sector from the technological characteristic $P = \varphi(U_{eff})$ of discharge; variation of inclination *D* (**a**) and intercept *C* (**b**) in the region of investigation.

This allows building the technological characteristics that correspond to the two operating sectors of the discharge external characteristic. The main characteristics, inclination *D* and intercept *C*, for the first technological regime of treatment are shown in graphic form in Fig. 4, and the main characteristics for the second technological regime of treatment in tabular form, Tables 2 and 3.

Table 2. Main characteristics of the second operating sector from the technological characteristic $P = \varphi(U_{eff})$ of the discharge - variation of inclination *D* in the region of investigation.

Second operating sector		Thickness of glass barrier δ, mm					
		3.0	**4.0**	**5.0**	**6.0**	**7.0**	**8.0**
Size of gap *b*, mm	**1.50**	1.2618	1.2366	1.2768	0.5136	-	-
	3.00	1.3418	1.3669	1.2805	1.3958	0.7893	-
	6.00	3.3008	-	1.7366	1.3391	1.2576	1.1271
	9.00	2.5062	-	5.1760	2.6556	3.6648	0.7009
	12.00	0.5832	-	9.5389	0.3743	0.2374	1.1832
	15.00	0.4803	-	-	0.4302	-	-
$P = \varphi(U_{eff})$		Inclination *D*, W/kV					

Table 3. Main characteristics of the second operating sector from the technological characteristic $P = \varphi(U_{eff})$ of the discharge - variation of intercept *C* with in the region of investigation.

Second operating sector		Thickness of glass barrier δ, mm					
		3.0	**4.0**	**5.0**	**6.0**	**7.0**	**8.0**
Size of gap *b*, mm	**1.5**	-11.0459	-12.1222	-15.8183	-4.3390	-	-
	3.0	-11.7055	-13.4985	-14.9070	-17.4430	-9.02606	-
	6.0	-44.3485	-	-22.7515	-18.0766	-18.2896	-16.2746
	9.0	-45.6125	-	-96.2239	-48.7976	-73.4814	-11.2244
	12.0	-8.2814	-	-199.8381	-5.2884	-3.2461	-23.6695
	15.0	-7.5314	-	-	-	-	-
$P = \varphi(U_{eff})$		Intercept *C*, W					

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