

THE CATHODE LAYER CHARACTERISTICS OF THE NORMAL DC ATMOSPHERIC PRESSURE GLOW DISCHARGE

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Abstract. The normal cathode fall and current density of the atmospheric pressure glow discharges for the different “cathode material – working gas” pairs are determined. The gas temperature in the cathode region was measured in each case. Laws of similarity were used to establish the conformity of obtained data to the corresponding ones for the low pressure glow discharges.

1. INTRODUCTION

There is large increasing interest in Atmospheric Pressure Glow Discharges (APGD) because they can be used for a wide range of technological applications without the need of vacuum systems. Some fundamental properties of the APGD plasmas have been experimentally characterized including discharge dynamics, optical emission, and densities of charged particles and excited species, but the experimental cathode fall parameters of the APGD have not been determined. To a lesser extent the APGD have also been studied numerically. Results of these numerical studies agree favorably with the macroscopic features of the measured discharge current and voltage. Theoretical models offer useful tools to understand atmospheric glow discharges, but precision of model results isn't high due to an imperfect data of elementary processes rates, especially, large uncertainties in the electron yield per ion for practical cathodes. That is why the results of every model calculation need experimental testing.

There are a lot of experimental data relatively the normal cathode fall and current density in the Low Pressure Glow Discharges (LPGD) presented in well-known books [1-4], for example. The dependence of the normal cathode fall of the LPGD on pressure was discovered in [5]. It was established that the cathode fall V_c decreases on 10÷15% at the increase of pressure from 1 Torr to a few tens of Torr in the LPGD in helium and neon with steel cathode. At the same time the change of cathode fall in the neon LPGD with titanium cathode was not observed. As to

atmospheric pressure, there are a few references where the experimental cathode fall parameters of the APGD are presented. The increase of the gas pressure up to the atmospheric one leads to a decrease in the dimensions of the glow discharge region characteristics and to the sharp increase in the heat release in cathode region. Determination of the normal cathode fall and current density of the different APGDs and their testing using laws of similarity are important current research topics and necessary for further optimization of the different APGD applications.

The cathode fall parameters were investigated in details for the self-sustained normal dc APGD in helium with the stainless steel cathode in [6]. The objectives of this work are to determine both the cathode fall and current density in self-sustained normal dc APGDs in other gases, namely, argon, neon, nitrogen, air and carbon dioxide. At the same time the different cathode materials are used as well.

2. EXPERIMENT

The experiments were performed in the installation geometry described in [6]. The electrodes were put in a pressurised chamber. The weakly rounded tungsten anode (6 mm in dia) was used in our experiments. To the contrary, the different material planar water-cooled cathodes were used. A working gas (helium, argon, neon, nitrogen, air and carbon dioxide) at a flow of 2 litre/min at atmospheric pressure was provided through the discharge chamber. The glow discharge was ignited by contacting anode and cathode and then by moving away one of each other. The interelectrode gap was about 2 mm.

Table 1. Current regions of the self-sustained normal dc APGD burning (Ampere)

Gas Cath. material	He	Ne	Ar	N ₂	Air	CO ₂
Copper	0.02 – 8	0.02 – 1.6	0.01 – 2	0.06 – 1.1	0.1 – 0.4	0.02 – 0.1
Stainless steel	0.02 – 6.5	0.02 – 1	0.01 – 0.3	0.08 – 0.6	0.1 – 0.4	0.02 – 0.1
Titanium	0.08 – 1	0.03 – 0.6	0.03 – 0.1	0.02 – 0.1		
Duralumin	0.01 – 0.2	0.01 – 0.1				

The discharge was fed by controlled dc power supplies with output voltage up to 600 V and 1200 V. The ballast resistance in the anode circuit was changed from ~75 Ω up to 2.5 kΩ. The voltage applied to electrodes and the discharge current were measured by means of a digital dc voltmeter (CH300, instrumental error 0.1%) and an amperemeter (M1104, instrumental error 0.2%), correspondingly. In table 1 the current ranges for different pairs of the gases and cathode materials where the APGD was observed in given experiments are presented.

The photo of the APGD at discharge current of 1 A in helium is presented in fig. 1. One can see, the structure of the glow discharge is as follows: a thin (less than 1 mm thick) disc of glow emission resides above the cathode surface, whereas the glowing column is adjacent to anode.



Fig. 1. Photo of the APGD in helium.

There is the Faraday dark space between these glow regions. The anode surface is covered with a glowing layer. The positive column is constricted to a diameter about 3–5 mm.

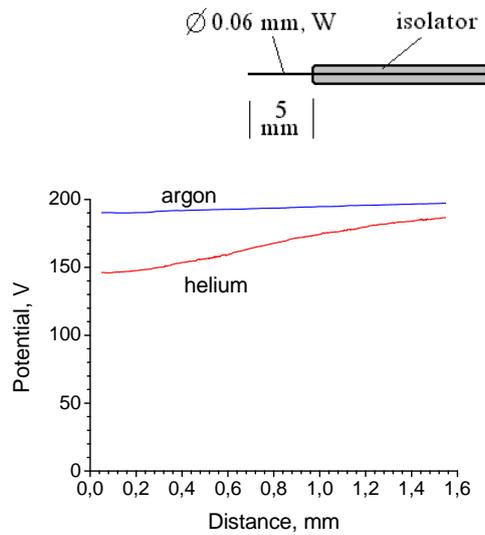


Fig. 2. Axial profiles of potential in the APGD in helium and argon at discharge current of 0.5 A.

The electric probe technique was used for the cathode fall measurements. The cathode fall voltage V_c was measured with the help of a tungsten probe $60 \mu\text{m}$ in diameter (see an upper picture insert in fig. 2).

The length of the uninsulated wire was 5 mm. The probe was put into discharge from one side and it was parallel to the cathode surface. The probe could move along the discharge axes by a stepper motor and by a microscrew in perpendicular direction.

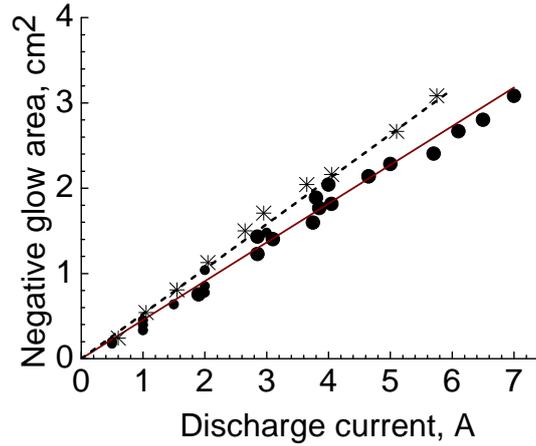


Fig. 3. Dependence of the area of the negative glow S on discharge current for the APGD in helium: closed circles – copper cathode, snowflakes – stainless steel cathode.

The probe potential V_p was measured by a digital voltmeter CH300 (input resistance is 10 M Ω) and determined by two ways. In most cases it was measured with respect to the grounded cathode. We supposed that $V_c \approx V_p$ at the instant just before the probe touched (or just after it detached) of the cathode. Axial profiles of potential in the APGD in helium and argon at discharge current of 0.5 A are presented in fig.2 for example.

In the cases when the probe setting into cathode region brought to the discharge instability the other way was used to determine the cathode fall. In this case the probe potential V_p was measured with respect to anode. At the same time the interelectrode voltage V_{ac} was determined. The cathode fall is $V_c \approx V_{ac} - V_p$ at the small interelectrode gaps in ranges of 0.2 – 0.5 mm.

The current density was defined as the ratio of discharge current I to the area of negative glow S . In fig. 3 dependences of negative glow area S on discharge current for the APGD in helium with the copper and stainless steel cathodes are presented. The linearity of the dependence of negative glow area on discharge current indicates that current density is constant, as in the case of a normal glow discharge. According to the figure 3 current densities are 2.3 A/cm² and 1.8 A/cm² for copper and stainless steel cathodes correspondingly. It is necessary to note that the water-cooling of cathode was applied at discharge currents more than 0.5 A to provide normal current density. The measurements were mainly fulfilled at the

discharge current of 0.2 A. But we always carried out the experiments at the range of discharge current lower and higher of this value to make sure that it is a normal mode of the APGD.

As it is known, current density in cathode fall depends strongly on the gas temperature in this volume even at discharge current of the order of 1 mA. More definitely, it depends on a gas density. Therefore, knowledge of the gas temperature is necessary for every measurement of current density to use the laws of similarity. The method of the relative rotational line intensity of molecular gases was used for the gas temperature determination [7-9]. The band (0,1)($\lambda = 427.81$ nm) from the first negative system of molecular nitrogen ion $N_2^+(B^2\Sigma_u^+)$ served as a working band. When this band wasn't observed (for example in case of argon or neon) we used (0, 0) ($\lambda = 308$ nm) OH band to determine the gas temperature.

3. RESULTS

The normal cathode falls V_c for the LPGD and APGD are given in table 2. Cathode falls we measured in the APGDs for various “cathode material – working gas” pairs are shown in thick print lines. The pressure value of 740 Torr is typical atmospheric pressure for Minsk (Belarus) where experiments were fulfilled.

Table 2. The normal cathode falls in the APGDs

Gas Cathode material	He		Ne		Ar		N ₂	
	<i>P</i> , Torr	<i>V_c</i> , V	<i>P</i> , Torr	<i>V_c</i> , V	<i>P</i> , Torr	<i>V_c</i> , V	<i>P</i> , Torr	<i>V_c</i> , V
Copper	1	177 [1] 204 [5]	1	220 [1] 200 [3] 148 [5]	1	130 [1,3]	1	208 [1]
	740	145±5	740	145±5	740	190±5	740	202±5
Iron	1	150 [1,3]	1	150 [1,3]	1	165 [1,3]	1	215 [1]
Steel	20	131 [5]	40	129 [5]	740	185±5	740	195±10
	740	140±5	740	145±5				
Titanium	1	115 [1,3]	1	114 [1,3] 113 [5]	1	99 [1,3] 99 [5]	740	380±10
	740	142±5	740	155±5	740	150±5		
Aluminum	1	140 [1,3]	1	120 [1,3]	1	100 [1,3]		
Duralumin	3.5	<173 [10]	740	125±5				

As one can see in table 2, the normal cathode falls of the APGDs are differing from ones for the LPGDs in sufficient bulk of cases. The increase or decrease of the normal APGD cathode falls at the transition from low pressure to atmospheric one depends on the “cathode material – working gas” pairs.

The cathode fall values obtained by us for the APGD with the stainless steel cathode in helium, neon and nitrogen do not practically differ from ones in discharges at 1 Torr. The cathode falls V_c in the APGDs with the copper and stainless steel cathodes in helium and neon are significant less than in the corresponding LPGDs. At the same time the cathode fall in the nitrogen APGD with the same cathodes does not change. The significant growth of the cathode fall voltage with pressure increase takes place for both the copper and titanium cathodes in the argon APGD.

There are simple seeming explanations of observed differences, namely, the differences in both the cathode surface preparation and the gas composition in low and atmospheric pressure experiments. At low pressure the cathode cleaning by the high-current glow discharge was used when the sputtering of cathode material takes place. But the APGD itself is the high current glow discharge and the sputtering takes place in it. We used in the experiments the gases containing the impurities less than 0.02%. At the same time, our experiments [14] showed that the addition of the admixture of other gases into working gas less than 0.1% doesn't have effect on the electrical parameters of the APGD. Therefore, probably, the differences of the cathode fall values at low and atmospheric pressure are due to pressure effect.

Current densities in the cathode region of both the LPGD and APGD are presented in tables 3 and 4. Ibidem the corresponding temperatures in cathode region of the APGDs are given as well. According to references the temperature in cathode region of the LPGD at pressure 1 Torr is 290 K, i.e. it is a room temperature.

Table 3. The normal current density of the glow discharge. Rare gases

Cath. material	Gas	He			Ne			Ar		
		LPGD		APGD	LPGD		APGD	LPGD		APGD
		j , $\mu\text{A}/\text{cm}^2$	j , A/cm^2	T_{g_s} , K	j , $\mu\text{A}/\text{cm}^2$	j , A/cm^2	T_{g_s} , K	j , $\mu\text{A}/\text{cm}^2$	j , A/cm^2	T_{g_s} , K
Copper			2.3 ± 0.2	720		2.3 ± 0.2	650	40 /5/	3.8 ± 1.5	980
Iron		2.2 [1,3]			6 [1,3]			160 [1] 100 [11]		
Stainless steel			1.8 ± 0.2	650		1.2 ± 0.2	740		2.5 ± 0.4 [13]	550
Titanium			1.0 ± 0.1	600		0.8 ± 0.1	690		1.0 ± 0.2	910
Aluminum		<15 [10]						110± 20 [12]		
Duralumin			1.4 ± 0.1	615		1.1 ± 0.1	615			

Table 4. The normal current density of the glow discharge. Molecular gases

Gas Cath. material	N ₂			Air			CO ₂		
	LPGD	APGD		LPGD	APGD		LPGD	APGD	
	j , μA/cm ²	j , A/cm ²	T_g , K	j , μA/cm ²	j , A/cm ²	T_g , K	j , μA/cm ²	j , A/cm ²	T_g , K
Copper		8.0 ± 0.5	2700	240 [1]	11 ± 1	3000		11.5 ± 1	2800
Iron Stainless steel	400 [1,3]	5.0 ± 0.5	2100		6 ± 1	2400			

Current density in the LPGD with various “cathode material – working gas” pairs changes on two orders of magnitude. For the LPGD with stainless steel cathode, for example, current density is 2.2 μA/cm² in helium discharge and 400 μA/cm² for nitrogen one. In case of the APGD the current density changes only on order of magnitude, namely, from ~ 1 A/cm² up to ~ 10 A/cm². Current density in both the LPGD and APGD is high in the molecular gas discharges in comparison with ones in rare gases. However the difference between the APGD current densities for various “cathode material – working gas” pairs is less than in case of the LPGD. It can be explained by a strong gas heating in discharges with high current density. Gas temperature at the end of cathode region in rare gas APGD does not exceed 1000 K. At the same time the gas temperature is in the range of 2000–3000 K in the molecular gas APGD.

4. LAWS OF SIMILARITY

Gas discharge parameters at different gas densities are connected by laws of similarity [1]

$$V=V_b, \quad d = d_l \frac{p_l T}{p T_l}, \quad j = j_l \left(\frac{p T_l}{p_l T} \right)^2, \quad (1)$$

which are correct when a number of limitations on discharge ionization mechanisms are fulfilled. Here d – cathode fall thickness. Parameters with subscript l correspond to the low pressure discharge. But laws of similarity (1) can not be used immediately for the comparison of the LPGD and APGD parameters due to gas temperature change in the APGD cathode layer. It takes place due to significant volumetric heat release. In this case it is necessary to use the one-dimension models of cathode fall region taking into account a volumetric heat release [15, 16]. These models, essentially, are a summarizing of similarity principle on a gas with inhomogeneous density.

The model of [15] is based on the following assumptions: (i) gas heating in the cathode fall is caused by collisions between the positive ions and neutral gas atoms; (ii) both the electric field and the ion current density decrease linearly with distance from the cathode; (iii) gas thermal conductivity is proportional to gas

temperature, $\lambda(T) = \alpha T$. In this model, the heat conduction equation for the gas in the cathode fall has the form

$$T \frac{d^2 T}{dx^2} + \left(\frac{dT}{dx} \right)^2 + \frac{E_{\max} j}{\alpha} \left(1 - \frac{x}{d} \right)^2 = 0, \quad (2)$$

where j is current density, E_{\max} is electric field near the cathode surface, and d is the cathode fall thickness.

Then according to [15], the cathode fall region, which is nonuniform in temperature, is replaced by a sheath with a uniform average temperature $\langle T \rangle$, and the electric parameters of the cathode fall are determined by using laws of similarity for a normal glow discharge (1). Expression for gas temperature within a cathode layer obtained in [15] is approximate. It is correct in asymptotic approximation of very high heat release. Exact solution of equation (2) has a following form [6]:

$$T = T_c \sqrt{1 + q(1 - \xi^4)}, \quad (3)$$

where $\xi = 1 - x/d$. Then

$$\langle T \rangle = T_c \left[\frac{1}{3} + \frac{\sqrt{2}}{3} \sqrt{1+q} \sqrt[3]{1 + \frac{1}{q}} F(\varphi, \frac{1}{\sqrt{2}}) \right]. \quad (4)$$

Here $F(\varphi, k)$ is the elliptic integral of the first kind and

$$\varphi = \arcsin \frac{\sqrt{2}}{\sqrt{1 + \sqrt{1 + (1/q)}}}, \quad (5)$$

where the dimensionless parameter

$$q = \frac{V_c j d}{3 \lambda_c T_c} \quad (6)$$

is the heat release power in the cathode fall normalized to the heat flux with a gradient near the cathode surface of $2T_c/d$. Here, V_c is the cathode fall voltage and λ_c is the gas thermal conductivity at the temperature T_c . The exact solution of the heat conduction equation [6] was used at calculations according to model [15] (model ESS).

In [16] (model BS), an attempt was made to calculate the parameters of the cathode fall with allowance for a nonuniform temperature distribution in it. According to [16], the local characteristics of the cathode fall at atmospheric pressure can be expressed in the form

$$dx = \delta(x) \frac{p_l T(x)}{p T_l} dx_l, \quad E(x) = \varepsilon(x) \frac{p T_l}{p_l T(x)} E_l(x_l), \quad (7)$$

where $\delta(x)$ and $\varepsilon(x)$ are the corrections related to the deviation of the current density j from the normal local value $j_n(x)$ determined from the scaling law. To calculate the corrections $\delta(x)$ and $\varepsilon(x)$, we used formulas

$$\frac{j}{j_n(x)} = \frac{1}{\delta(x)(1 + \ln \delta(x))^2}, \quad \varepsilon(x) = \frac{1}{1 + \ln \delta(x)}. \quad (8)$$

The temperature profile in the cathode fall is calculated from the heat conduction equation

$$\frac{d}{dx} \left(\lambda(T) \frac{dT}{dx} \right) + E j_T = 0, \quad (9)$$

where j_T is the fraction of the current density that causes gas heating. For a normal glow discharge, the current density j is believed to correspond to the minimum of the cathode fall voltage. So, it is necessary to find such solution of a boundary problem of the heat conduction equation added by algebraic relation, which minimizes of the cathode potential of cathode layer. In [16], in contrast to [15], it was the electron current also contributes to the cathode fall along with the ion current.

5. DISCUSSION

Let's compare the parameters of cathode fall region using the one-dimension models [15, 16] of this region. According to model [15] the cathode fall does not depend on gas pressure. At the same time the cathode fall determined by model [16] increases while pressure is increasing. The stronger temperature inhomogeneity in cathode layer is the more this increase is. However even in case of the molecular gases the difference of cathode fall values in glow discharges at atmospheric and low pressures does not exceed a few Volts. Therefore, the significant differences of cathode fall values at low and atmospheric pressure marked above (see table 2) can not be explained in frames of models [15, 16].

Experimental and calculated according to models [15, 16] values of both the current density in cathode region and gas temperature in negative glow are presented in tables 5 and 6. The cathode fall region parameters corresponding to pressure of 1 Torr [1] were used in these calculations.

Table 5. Experimental and calculated parameters of the cathode fall. Rare gases

Gas-cath. material	He-Fe		Ne-Fe		Ar-Fe	
	j , A/cm ²	T , K	j , A/cm ²	T , K	j , A/cm ²	T , K
Experiment	1.8±0.2	650	1.2±0.2	740	2.5±0.4	950
Model ESS	0.47	470	1.0	540	6.5	1230
Model BS	0.48	465	1,1	520	7.4	1240

Table 6. Experimental and calculated parameters of the cathode fall. Molecular gases

Gas-cath. material	N ₂ -Fe		Air-Cu	
	j , A/cm ²	T , K	j , A/cm ²	T , K
Experiment	5.0±0,5	2100	11.0±1.0	3000
Model ESS	8.7	1730	7,7	1400
Model BS	6.9	2280	6,2	1850

The calculated current density for steel-helium pair is less than experimental one in 4 times. Calculated gas temperature is below as well. The opposite situation is in the APGD in argon when the calculated current density value exceeds the experimental one in a few time, and calculated temperature is significantly high than experimental one. A good agreement between the calculated and experimental values takes place for discharges in neon and nitrogen. Probably, the observed differences between the calculated and experimental data are significant above all about changes in the elementary process balance in the discharge plasma at the transition from low pressure to atmospheric one.

In [6] a comprehensive investigation of the APGD in helium with stainless steel cathode was fulfilled. Besides a cathode fall, current density and gas temperature the cathode fall thickness and heat flux to the cathode were determined as well. A good agreement between experimental and calculated values including a heat flux to the cathode was observed. However it was necessary to use the increased value of current density (in 6 times) and the increased cathode fall thickness (in 2 times) in comparison with classical LPGD parameters at these calculations. Experimental determination of the three main parameters of the cathode fall region (V , j and d) allows, probably, to calculate more exactly a heat mode of discharge operation in cases of other gases. The last is very important for a number of applications.

6. CONCLUSIONS

The normal cathode fall and current density of the atmospheric pressure glow discharges for the different “cathode material – working gas” pairs were determined. The gas temperature in the cathode region was determined in each case. The laws of similarity were used to establish the conformity of obtained data to the corresponding ones for the LPGDs.

The calculated current density for steel-helium pair is less than experimental one in 5 times. The opposite situation is in the APGD in argon when the calculated current density value exceeds the experimental one on the order of magnitude. A good agreement between the calculated and experimental values takes place for discharges in neon and nitrogen. Probably, the observed differences between the calculated and experimental data are significant above all about changes in the elementary process balance in the discharge plasma at the transition from low pressure to atmospheric one.

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