Investigation of changes in field electron emission characteristics of industrial fine-grained graphite when operated in an argon atmosphere up to $10^{-2}$ Pa

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Abstract. Studying of field electron emission properties of carbon cathodes operating under technical vacuum conditions is a promising scientific field. Massive cathode made of commercial fine-grained graphite of MG (Russian abbreviation) grade is being investigated. Experiments on obtaining current-voltage characteristics and long-term testing are being carried out. The emitter made of fine-grained graphite demonstrates good emission properties under technical vacuum conditions. Carbon cathode is capable of operating at pressures up to $2\times10^{-2}$ Pa. Increased pressure in the vacuum chamber leads to deterioration of cathode emission properties. Electric field enhancement factors were calculated for all stages of studies. Analysis of experimental data demonstrates decrease in enhancement factor due to ion bombardment of cathode surface during exploitation. This results in higher electric field for operation of investigated graphite cold cathodes.

1. Introduction
One of the popular fields of vacuum electronics nowadays is development of field electron emission cathodes. Most of the investigations are dedicated to nanostructured emitters: carbon nanotubes (CNT) [1, 2], metal nanotubes [3, 4], semiconducting nanoparticles [5-7]. However, high current, Joule heating, and bombardment with ions of residual gases destruct emitters, decrease cathode lifetime, and limit current of an individual nanotube to 1 $\mu$A [1]. A majority of the described nanoemitters produce field emission current only of the microampere range [1, 3-7] whereas industrial vacuum devices — such as electron guns and X-ray tubes — require reliable cathodes with the current of several milliampères and greater. Another disadvantage of nanostructured cathodes is a huge threshold electric field that reaches tens of kilovolts per millimeter [4-7]. Therefore, semiconducting nanoemitters seem more suitable for development of field effect transistors and devices for photovoltaic applications. Moreover, it is challenging to produce tubes with desired properties and provide appropriate electrical contact with electrode [3]. Authors studied field electron emission cathode made of massive graphite produced for iron and steel industry. Fine grained graphite MG (Russian abbreviation) was chosen for the experiments. Such type of materials offers the following essential advantages: it does not require special preparation, can operate under technical vacuum conditions (pressure of $10^{-5}$–$10^{-7}$ Pa, intense ion bombardment), generates field emission current of the milliampere range [8], and is cost-efficient. Resistance to ion bombardment is important, for residual gases always can be found in vacuum devices. From the above, this investigation is relevant to development of cold-emitter devices, and
massive graphite is considered a promising material for such type of cathodes due to low work function, developed surface with numerous emission centers, as well as high melting point, and heat and radiation resistance [9, 10].

Understanding field electron emission processes in vacuum electronics is important because even in the case of traditional cathodes, field emission takes place for other elements of the system operating in strong electric fields. Investigation of the properties of carbon materials is also relevant, since these materials are widely used in vacuum devices.

Stable operation of nanostructured emitters requires the condition of ultrahigh vacuum. But massive cathodes due to weaker dependence on the efficiency of single emission centers can function even under technical vacuum conditions, at a pressure of more than $10^{-4}$ Pa [11]. Since vacuum devices operate in an atmosphere of residual gases, the device elements are exposed to ions and molecules of these gases. The transformation of the materials characteristics under such influences can lead to breakdowns, incorrect device operation and its failure. Taking these processes into account will improve the efficiency and reliability of the developed vacuum devices.

2. Materials and equipment

The sample explored (MG) is a kind of fine-grained graphite. The cathode has the following key properties: electrical resistivity is $13 – 18 \ \mu\Omega\cdot m$, ash content is $0.5\%$, typical grain size is $0.5$ mm, and porosity is $30\%$. This material is used for manufacture of screens, melting pots, electrodes, etc. exploiting under high temperature in metallurgical industry. Material from the group of grain structure graphite cathodes was chosen due to the best combination of emission and mechanical properties demonstrated in the previous stages of research: low operating voltage, high stability, mechanical strength and ease of processing [11].

Experiments were carried out on the installation described in [12]. Figure 1 shows its block diagram. The cathode has a cylindrical shape (a round rod), $4$ mm in diameter, and is mounted in a holder. The anode is made of carbon. The electrode system employs plane-parallel geometry in order to generate a homogeneous electric field in the gap. Such a system, cathode and anode, is hereinafter referred to as “a measuring (or experimental) cell”. A high voltage source (HVC) generates an electric field to initiate field electron emission. This device is controlled by computer (PC) via input-output system RL-88AC. The last collects experimental data from the registration system: a voltage divider and a resistive shunt.

![Figure 1. Block diagram of experimental installation: HVS – high voltage source; PC – personal computer.](image)

The interelectrode gap was set to $0.5$ mm. The typical pressure level is $2 \times 10^{-4}$ Pa. Experiments of two types were carried out. The first is obtaining current-voltage characteristics (CVC). The chart obtained shows the relation between emission current and macroscopic electric field strength $E$. The last is defined as

$$E = \frac{U}{d},$$

(1)
where $U$ is anode voltage, $d$ is the gap between electrodes.

The second type of experiments is long-term testing (LTT): emission under current stabilization during long time. Long-term experiments reflect how the properties of experimental cell change over a period of tens of minutes. Such an approach allows finding out whether the cathode degrades, works stably, or improves its emission properties.

Argon was chosen to study the field electron emission properties of carbon materials in a gas atmosphere. It is a gas traditionally used in vacuum technology for various tasks: it is widely used, the mass of ions is sufficient to generate radiation effects when interacting with a surface, but does not cause critical damage.

3. Results and discussion

For the selected sample, a set of experiments was carried out: initial current-voltage characteristics (CVC) acquisition, long-term testing (LTT) at stabilization of the emission current at levels of 0.1, 0.2, and 0.3 mA. The same series of studies were performed when argon was injected into the vacuum chamber to a pressures of $6 \times 10^{-4}$, $2 \times 10^{-3}$, $6 \times 10^{-3}$, and $2 \times 10^{-2}$ Pa. For the convenience of description, each series of experiments at pressures of $2 \times 10^{-4}$ (maximum system pumping), $6 \times 10^{-4}$ (in argon), $2 \times 10^{-3}$ (in argon), $6 \times 10^{-3}$ (in argon) and $2 \times 10^{-2}$ Pa (in argon) are hereinafter denominated stages 1-5 correspondingly. Stage 1 is also referred to as “in vacuum”.

3.1. Current-voltage characteristics

CVC were registered both for the best vacuum conditions and for operation in an argon atmosphere. The former provides information on the intrinsic emission properties of the cathode, whereas the dependences of the current on the electric field at an increased gas pressure demonstrate the characteristics of the emitter in a given operating mode.

Figure 2 (a) demonstrates typical dynamics of CVCs after series of experiments for the cold cathode made of MG structural graphite. The characteristics were recorded after the emitter operation for a long time (10–20 minutes) at each given pressure level, which allows observing the change of cathode properties during operation under various conditions. After each LTT with an inlet of argon, CVCs in a gas atmosphere were obtained. After that, the characteristics at the best vacuum were registered.

![Figure 2. CVCs for MG sample: a – conventional appearance; b – Fowler-Nordheim plots.](image)

MG takes an intermediate position between grained graphite cold emitters. The range of the operating voltage required for the MG cathode is 2.5–9 kV, which corresponds 5–18 kV/mm. The smallest values of electric fields initiating field electron emission are typical for graphite with low ash
content (4-13 kV/mm). The maximum voltages are required for the emitter made of dense fine-grained graphite (7-20 kV/mm).

With an increase in the pressure level in the vacuum chamber, the CVCs of carbon cathode shift to the region of high electric fields. In this case, in the vast majority of cases, the characteristics obtained in vacuum correspond to lower operating voltages than CVCs recorded in argon atmosphere. Figure 2 (b) shows the Fowler-Nordheim plots for the same experimental data.

3.2. Field enhancement factor analysis
For all types of experiments performed, there is a tendency to an increase in the electric field strength required for the exploitation of carbon cathodes under conditions of increased operating pressure. The authors believe that this is achieved not only by worsening the conditions for the operation of the field emitter, but also by changing the properties of the cathode under increased residual gas pressure. In order to test this hypothesis, the analysis of the change in the electric field amplification factor of the cathodes was carried out. For the calculation, the formula (2) obtained from the original Fowler-Nordheim equation, taking into account the refinements and simplifications proposed in [13] was used,

\[ j = 1.4 \times 10^{-7} \cdot \frac{\beta^2 E_0^2}{\varphi} \cdot \exp\left[ \frac{10.11}{\varphi^{0.2}} \right] \cdot \exp\left[ -6.49 \cdot 10^7 \cdot \frac{\varphi^{0.2}}{\beta E_0} \right] , \]  

where \( \beta \) is a field enhancement factor, \( E_0 \) is a macroscopic electric field in V/cm (\( E_0 = U/d \)), \( \varphi \) is a work function of the cathode in eV and \( j \) is the current density in A/cm².

Taking into account that the total cathode area with a sample diameter of 0.4 mm is 0.126 cm², the work function of graphite is 4.7 eV [14], we obtain an expression for calculating the field amplification factor (3)

\[ \beta = -(6.61 \cdot 10^9 \cdot A^{-1}) , \]  

where \( A \) is the slope angle of Fowler-Nordheim plots.

Figure 3 shows the dynamics of the electric field enhancement factor \( \beta \), obtained from the results of experimental data processing.

Several conclusions can be drawn from the analysis of the data obtained. First, a decrease in the value of \( \beta \) in the experiments for the sample is observed. This is most likely due to the influence of ion bombardment during the cathode operation on the carbon surface state. Second, in the vast majority of cases, the value of \( \beta \)-factor obtained in the study of the cathode properties in an argon atmosphere is...
less than in the experiment in vacuum. Probably, intensive ion bombardment transforms the morphology, destroys the cathode surface, including the microtips – emission centers. This worsens the conditions for field electron emission and provides CVCs at increased voltage on the electrodes. This calculation helps to explain the deterioration of field emission properties of the carbon cathode at increased residual gas pressure.

4. Conclusions
Thus, it can be concluded that the injection of argon into the vacuum chamber up to the level of $2 \times 10^{-2}$ Pa leads to a deterioration in the emission properties of massive carbon cathode, in particular, granular structural graphite of MG grade. This effect is achieved due to bombarding the emitter surface with argon ions. Ion bombardment results in a transformation of the relief of the cathode surface layer. This factor, as shown in current work, causes a decrease in the average electric filed enhancement factor of the cathode. A reduction of this parameter leads to a decrease in the emission current at a given electric field or requires an increased operating voltage in order to achieve or maintain the desired current level. This feature can be used in vacuum devices to increase the electric strength of the interelectrode gaps, allowing operation at higher residual gas pressures. The application of carbon field emission cathodes makes it possible to develop vacuum devices that can operate in harsh vacuum conditions and withstand emergency operating modes.

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