# Development of the High-Voltage Electrodischarge Method for Obtaining Carbon Nanomaterials from Carbon-Containing Gases and Liquids

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Electrodischarge processing of carbon-containing gases is widely used to obtain carbon nanomaterials of various structures and purposes. This method has a number of disadvantages, the main of which is that when a certain power of the electric discharge is reached and/or its constant increase, the productivity grows more slowly or stops growing altogether, which leads to an increase in specific energy consumption per mass unit of the output product. Also, with an increase the length of the working gas gap, in which the gas is processed by plasma, the power of the electric discharge installation for obtaining nanomaterials can be increased. This effect is shown in [1] and illustrates in fig. 1 ($K^*_P$ is relative productivity, $l^*_{ed}$ – relative interelectrode gap and $P^*_{out}$ – relative output power.). However, the discharge gas gap cannot be more than 30-40 mm due to the fact that the breakdown voltage of a gap will be 60-80 kV, which can damage the equipment and is unsafe. The instability of the process also increases due to fluctuations in the length of the plasma column caused by the flow of gas inside the reactor.

Therefore, the **aim** of this work is to find a way to increase the productivity of carbon nanomaterials at a constant specific power consumption of high-voltage installations for the electric discharge treatment of carbon-containing gases without increasing the length of the interelectrode gap.

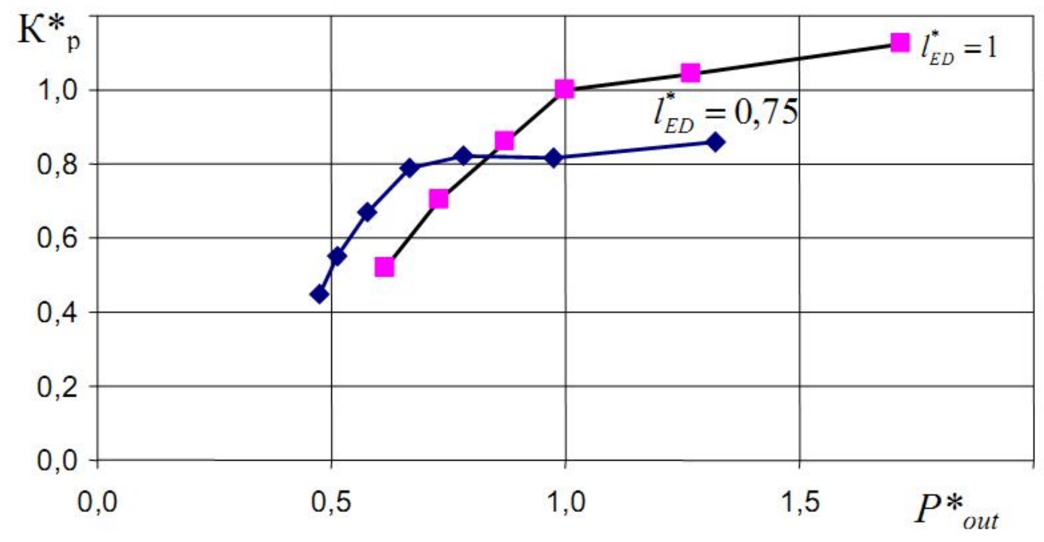

Fig. 1. Productivity of electric-discharge installations versus output power

The **main idea** of the work is that in order to increase the productivity of carbon nanomaterials at a constant length of the interelectrode gap, it is necessary to increase the density of the treated substance in the reaction zone during the electrodischarge processing. This can be achieved either by increasing the pressure of the carbon-containing gas or by using a carbon-containing liquid. These two methods will be described in this work. This is evidenced by the fact that when treating denser (liquid) hydrocarbons with a high-voltage electric pulse method, with the same performance, the length of the interelectrode gap in liquid processing [1-2] can be up to 5 times less than in electric discharge gas treatment [1]. To increase the density of the carbon-containing gas substance in the reactor, at a constant volume and, preferably, temperature, it is necessary to increase its static pressure. Therefore, it is necessary to increase the pressure inside the reactor in order to increase the productivity of electric-discharge processing installations for carbon-containing gases at a constant interelectrode gap.

Therefore, to achieve the aim and verify the idea of work, two scientific and technical tasks must be solved. The first task is to create an experimental stand for high-voltage electric-discharge treatment of carbon-containing gases with the possibility of stabilization and regulation of pressure inside the reaction zone. The second task is to carry out experimental studies of the possibility of increasing the productivity of high-voltage electric-discharge processing installations for carbon-containing gases with a fixed length of the interelectrode gap.

**Experimental Installation.** The block diagram of the experimental installation for high-voltage electric-discharge processing of carbon-containing gases is shown in Fig. 2. It consists of: a reactor with a gas inlet system and output adjustable pressure valve, a fixed bottom electrode, a movable top electrode, an electromagnetic driver for the top electrode, a high-voltage adjustable resonant power source, and the main control system of the installation.

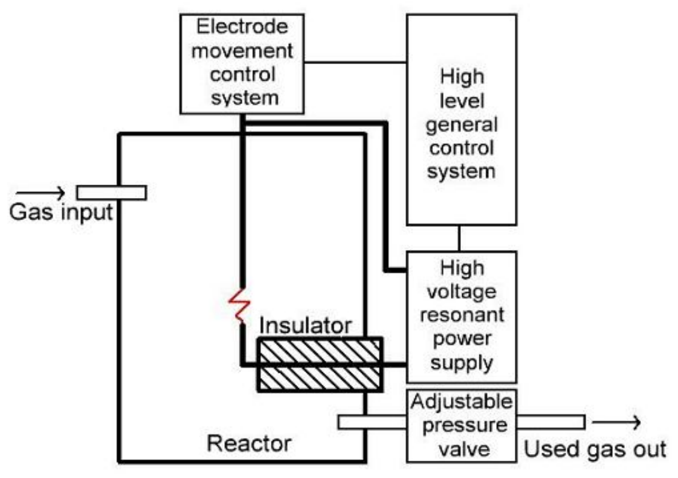

Fig. 2. Experimental high-voltage electric-discharge processing installation

An adjustable output pressure valve is needed to maintain a constant pressure in the reactor of carbon-containing gas, which is higher than atmospheric. The gas valve, safety membrane type KSV-451AC, produced in the Soviet Union, with adjustable pressure of 150 and 200 kPa with ±15 kPa error. As the input raw material for the production of carbon nanomaterials, the gas with the highest density of halogen-free carbon-containing gases - hydrocarbon butane was chosen. Gas is supplied directly from the balloon through the reducer. After the reducer, there is also a 250 kPa safety valve in case of contamination of the gas outlet from the reactor and preventing an increase in pressure in the reactor. The weight of the output product was measured with a laboratory balance scales with an accuracy of 10 mg.

When a high voltage is applied to the electrodes, a glowing electric discharge occurs between them, after the initial breakdown of the gas interelectrode gap, which serves as a tool for processing carbon-containing gas. The energy transferred by the power source to the electric discharge is spent on heating the gas in the interelectrode gap, its decomposition and the formation of new carbon nanostructures [3]. An electric discharge is powered by a high voltage high frequency resonant power source through the top and bottom electrodes. The bottom electrode is stationary. The top electrode has a mechanical drive for moving up and down. This is necessary in order to maintain the desired length of the interelectrode gap and to clear the interelectrode space of conductive carbon deposits. A high-voltage resonant power supply consists of a full-bridge voltage inverter, the output of which is loaded on a resonant circuit, which is formed by a set of resonant inductors and parasitic parameters of a high-voltage high-frequency transformer (transformation ratio is 60). The output current of the source is stabilized by the frequency-parametric method described in [4]. The inverter operates at a frequency of 16 to 60 kHz, the output current is from 40 to 200 mA RMS. Maximum output voltage of the power supply is 50 kV. There are current protection systems and a power factor corrector. The task of the high-level general installation control system is to maintain stable operation of the installation according to the algorithms described in [4]. The installation is built on the principles described in [4]. Some ideas can take from [5-8].

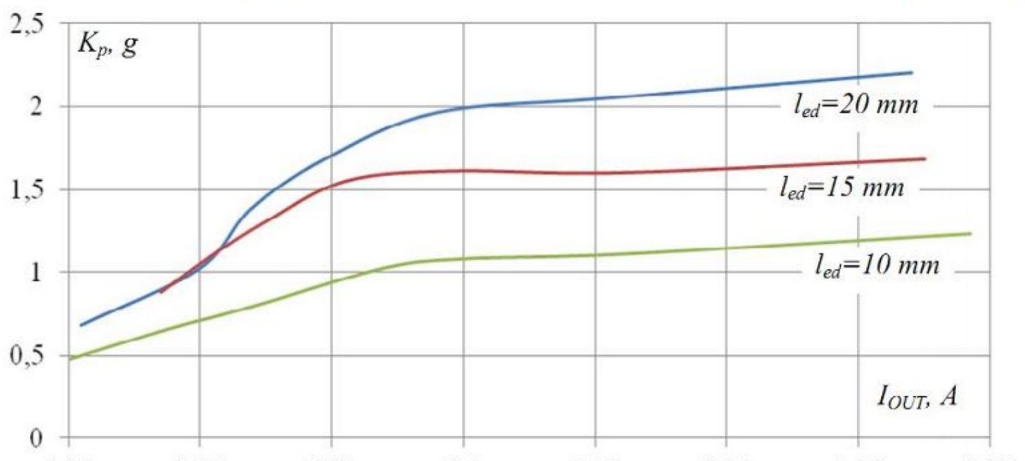

Fig. 3. Dependence of productivity on output current at atmospheric pressure

**Experimental Results.** Since the voltage on the discharge channel does not depend on the current through the channel in the mode of productive processing of carbon-containing gases [9], we can assume that the power in the discharge channel is proportional to the current through this channel. Therefore, for experimental studies of the dependence of productivity ($K_P$) on power, we will change the current of the discharge channel ($I_{OUT}$). The length of the interelectrode gap ($l_{ed}$) and the gas pressure ($p_s$) in athe reactor will also vary. The values of the interelectrode gap length are chosen equal to 10, 15 and 20 mm, the pressure in the reactor will be changed from atmospheric to 200 kPa, with an intermediate value of 150 kPa. The experiment, at atmospheric pressure, we will conduct for all three lengths of the interelectrode gap, and at a variable pressure only for 20 mm, since with this interelectrode gap there will be maximum productivity. The duration of each experiment is 15 minutes. The results of experimental studies at atmospheric pressure are shown in Fig. 3. As can be seen from the obtained experimental data, there are such values of the discharge channel current at which the performance begins to grow more slowly. For example, for the interelectrode gap $l_{ed}$ = 20mm this is the current value $I_{out}$ = 100 mA RMS with a productivity of 2 g in 15 minutes. Above this value, increasing the current does not make sense, since energy losses per unit mass ($W_p$) of the obtained carbon nanomaterial begin to increase as shown in Fig. 4.

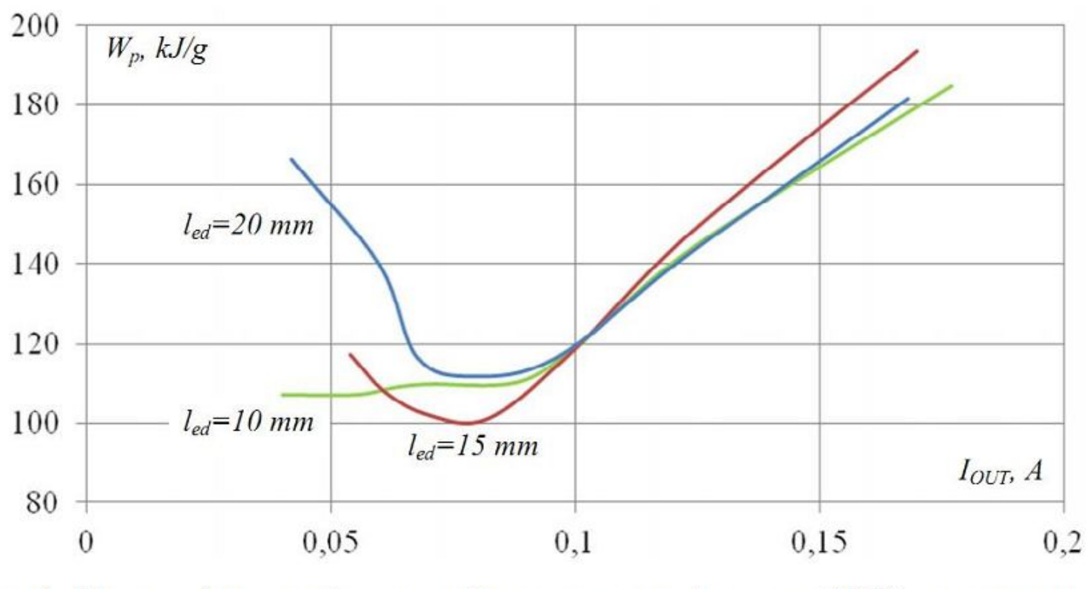

Fig. 4. Dependence of energy losses per unit mass ($W_p$) on output current at atmospheric pressure

Also, in Fig. 5, the dependence of the specific energy losses per unit mass on productivity is shown for various lengths of the interelectrode gap at atmospheric pressure in the reactor. The following conclusions can be drawn from the obtained experimental results: 1) for a fixed interelectrode gap, after reaching a certain productivity, the specific energy losses begins to increase rapidly (Fig. 5); 2) there is a range of optimal productivity to energy consumption for each value of the interelectrode gap; 3) to increase the productivity of electric-discharge processing installations for carbon-containing gases, with satisfactory specific energy losses, it is necessary to increase the length of the interelectrode gap. A further increase in the interelectrode gap is associated with insulation problems in the reactor, since the voltage of the initial guaranteed breakdown of the interelectrode gap of 20 mm long is 40 kV [9].

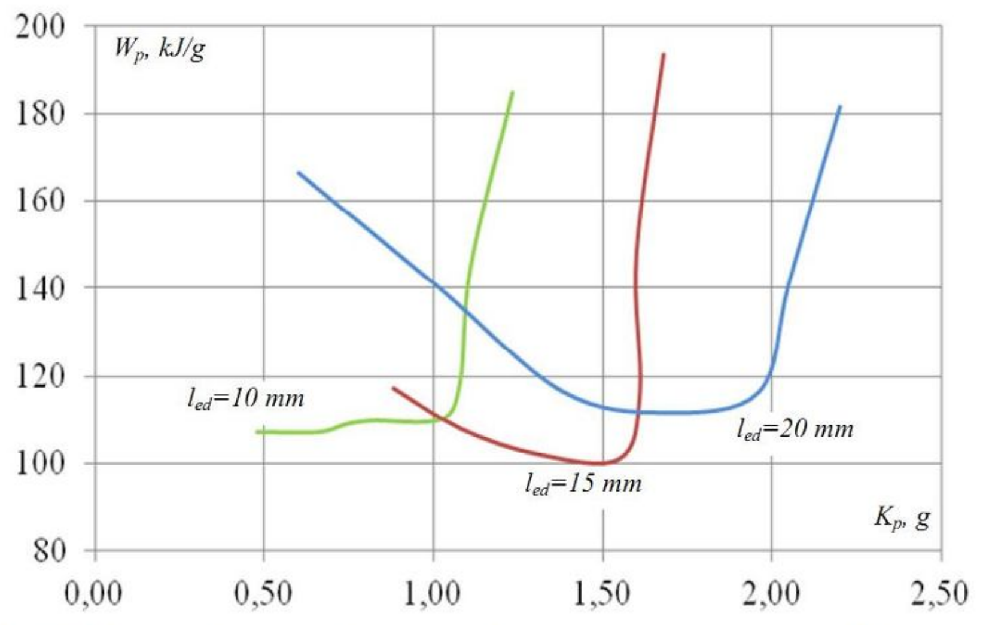

Fig. 5. Dependence of energy losses per unit mass ($W_p$) on productivity at atmospheric pressure

Therefore, the next way to increase productivity, with a constant length of the interelectrode gap, is to increase the pressure in the reactor and, as a result, increase the density of the carbon-containing gas in the reaction zone. The results of experimental studies of the effect of pressure in the reactor on the performance of electric-discharge treatment installations of carbon-containing gases, with an interelectrode gap of 20 mm and a change in the discharge channel current, are shown in Fig. 6. It can be seen that with increasing pressure in the reactor, the productivity of carbon-containing gas processing high-voltage installations also increases. We will also analyze the specific energy consumption and productivity, as was done for atmospheric pressure. The results are shown in Fig. 7 and Fig. 8.

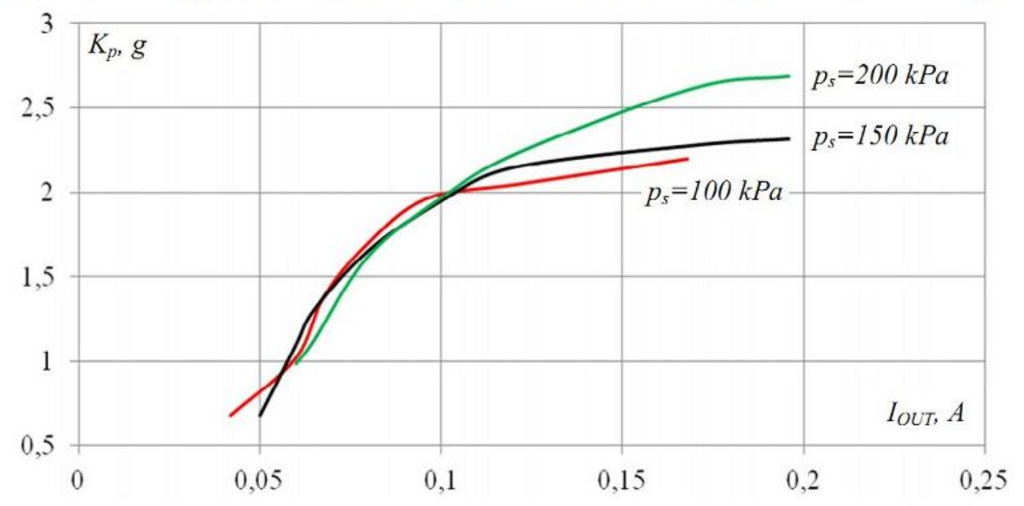

Fig. 6. Dependence of productivity on output current at various pressure (interelectrode gap of 20mm).

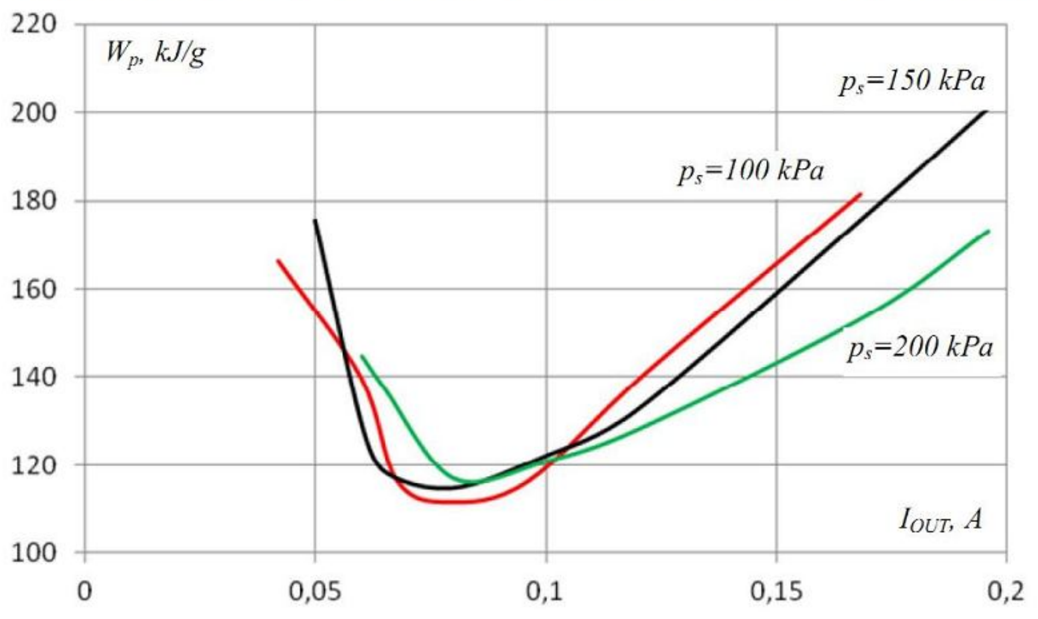

Fig. 7. Dependence of energy losses per unit mass on output current at various pressure

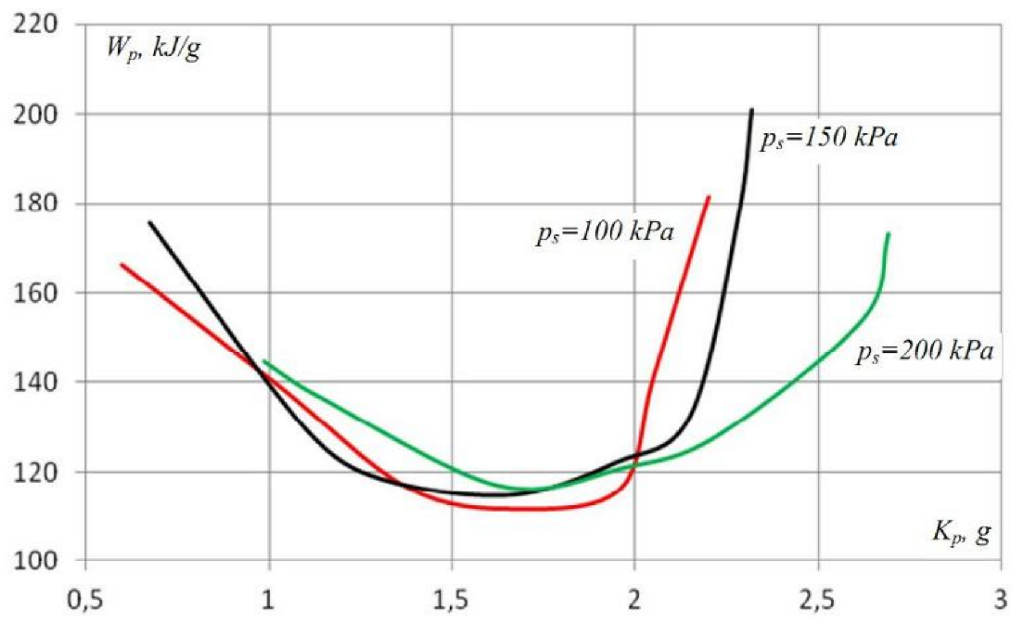

Fig. 8. Dependence of energy losses per unit mass on productivity at various pressure

The specific energy losses at elevated pressure are less than at atmospheric pressure, and productivity is greater. Experiments also show more stable plasma operation at elevated pressure. The increase in pressure shifts to the right in productivity without a significant increase in energy loss, the dependence of energy loss on productivity. Therefore, with increasing pressure, the productivity of electric-discharge carbon-containing gas processing installations increases without increasing the length of the interelectrode gap and, consequently, operating and start-up voltage. Therefore, it is advisable to increase the density of the processed raw materials in the reactor. Further research will be aimed at increasing the density of processed carbon-containing raw materials and the study of the possibility of electric-discharge treatment of carbon-containing liquids. The density of any carbon-containing liquid (for example, pentane or hexane) is orders of magnitude higher than that of gases (butane, propane) at atmospheric pressure and room temperature.

**Conclusion.** The work explores the possibility of increasing the productivity of high-voltage electric-discharge processing installations for carbon-containing gases without increasing the length of the working interelectrode gap by increasing the density of the processed carbon-containing raw materials. It is shown that with increasing pressure in the reactor, productivity increases without a significant increase in specific energy losses per unit mass of the output nanoproduct. It is advisable to increase the density of the processed raw materials in the reactor.

**References.**

[1] D. V. Vinnychenko. "Influence of the electrical parameters of high-voltage electrical discharge installations of nanocarbon synthesis on their productivity and specific energy consumption," (in Ukrainian), Technical Electrodynamics, no. 4, 2016, pp. 95-97.

[2] N. Kuskova, K. Dubovenko, S. Petrichenko, P. Tsolin and S. Chaban, "Electrodischarge technology and equipment to produce new carbon nanomaterials," Surface Engineering and Applied Electrochemistry, 2013, Vol. 49, Issue 3. – pp. 215-221.

[3] L. Z. Boguslavskii, A. D. Rud, I. M. Kir'yan, N. S. Nazarova and D. V. Vinnichenko, "Properties of carbon nanomaterials produced from gaseous raw materials using high-frequency electrodischarge processing," Surface Engineering and Applied Electrochemistry, vol. 51, no. 2, 2015, pp. 105-109.

[4] D. Vinnychenko and N. Nazarova. "The High-Voltage Electrical Engineering Systems of Gaseous Hydrocarbons Electro-Discharge Processing Design Principles " IEEE 39th International Conference on Electronics and Nanotechnology (ELNANO), 2019, pp. 678-683, doi: 10.1109/ELNANO.2019.8783220

[5] M. H. Rashid, Power Electronics Handbook (Second Edition). Devices, Circuits, and Applications. Elsevier Inc, 2007.

[6] A. A. Shcherba and N. I. Suprunovska, "Study features of transients in the circuits of semiconductor discharge pulses generators with nonlinear electro-Spark load," IEEE International Conference on Intelligent Energy and Power Systems (IEPS 2014), pp. 50-54, 2014.

[7] G. V. Pavlov, I. L. Vinnychenko and M.V. Pokrovskiy, "Adaptive control system of frequency converter on the basis of the resonant inverter with nonlinear control", (in Russian) Technical Electrodynamics, vol. 5, 2018, pp. 39-43.

[8] Shcherba A.A., Kosenkov V.M., Bychkov V.M. Mathematical closed model of electric and magnetic fields in the discharge chamber of an electrohydraulic installation. *Surface Engineering and Applied Electrochemistry*. 2015. Vol. 51 (6). Pp. 581-588.

[9] D. V. Vinnychenko. "Determination of optimal characteristics of the high-voltage electro-discharge system for the realization of the technology of electropulse synthesis of nanocarbon," (in Ukrainian), Technical Electrodynamics, no. 4., 2014, pp. 129-131.