Characteristics of cold atmospheric plasma source based on low-current pulsed discharge with coaxial electrodes

O A Bureyev, Yu S Surkov and A V Spirina
Institute of Electrophysics, Ural Branch of the Russian Academy of Science, 106 Amundsen St., Ekaterinburg, 620016, Russia
E-mail: bureyev@iep.uran.ru

Abstract. This work investigates the characteristics of the gas discharge system used to create an atmospheric pressure plasma flow. The plasma jet design with a cylindrical graphite cathode and an anode rod located on the axis of the system allows to realize regularly reproducible spark breakdowns mode with a frequency ~ 5 kHz and a duration ~ 40 μs. The device generates a cold atmospheric plasma flame with 1 cm in diameter in the flow of various plasma forming gases including nitrogen and air at about 100 mA average discharge current. In the described construction the cathode spots of individual spark channels randomly move along the inner surface of the graphite electrode creating the secondary plasma stream time-average distributed throughout the whole exit aperture area after the decay of numerous filamentary discharge channels. The results of the spectral diagnostics of plasma in the discharge gap and in the stream coming out of the source are presented. Despite the low temperature of atoms and molecules in plasma stream the cathode spots operation with temperature of ~ 4000 °C at a graphite electrode inside a discharge system enables to saturate the plasma by CN-radicals and atomic carbon in the case of using nitrogen as the working gas.

1. Introduction
The source under discussion belongs to the class of devices named in literature «Cold Plasma Jet». This term is used for sources of non-equilibrium atmospheric pressure plasma flow with temperature of heavy particles in the range of 300-1000 K that is at least one order of magnitude lower than the electron temperature in such plasma [1]. Application of these devices allows treatment of thermo-sensitive materials with simultaneous reduction of cost as compared with low-pressure plasma technologies as there is no need to use vacuum installations.

The results of works on modification of materials in cold atmospheric plasma [2,3] show that such treatment most often leads to simultaneous development of surface morphology (increase of roughness after plasma-chemical etching) and increase of hydrophilicity because of doping of polar functional atomic groups containing oxygen and nitrogen onto the surface. One of important advantages of cold atmospheric plasma treatment as compared with thermal treatment or electrolytic processes is modification of the only surface layer without mechanical or thermal damage of the material bulk. Energy consumption and environmental hazard of production process are reduced as well.

Characteristics of the source of a new type creating a jet of cold atmospheric plasma near 1 cm in diameter for different working gases including nitrogen and atmospheric air are studied in this work. The main feature of the source operation is the chaotic movement of cathode spots of low-current pulse discharge inside a cylindrical graphite cathode with the creation of secondary plasma flow after decay of numerous spark current channels.
2. Methods of experiment

Plasma source diagram is presented in figure 1. Discharge system with coaxial geometry contains a steel rod anode on the axis and a hollow tubular cathode made of isotropic graphite mounted at the end of quartz pipe through which the working gas is fed into the discharge gap under excess pressure. In the process of source operation, decaying plasma is forced out of the discharge gap by the flow of the working gas through the cathode aperture $\varnothing$ 8 mm, forming a plasma flame. Technically pure argon and nitrogen were used as working gases as well as atmospheric air of natural humidity pumped by a compressor.

Electric power for discharge system was supplied by a high voltage pulse power source with a pulse rate regulated in the range of 10 Hz - 7.5 kHz. To prevent the discharge from transition into the arc mode, ballast resistor 4 kΩ was installed into the discharge circuit. The amplitude of high voltage pulses of positive polarity was 5 kV. Pulses of open circuit voltage have rise time $\sim$10 $\mu$s, the time of descent being approximately 100 $\mu$s. The duration of discharge current pulses was 40 $\mu$s.

The measurement of average values of discharge current and discharge voltage was fulfilled by means of magneto-electric system devices. Pulse discharge current was measured by 1.3 $\Omega$ shunt. Low discharge currents were measured by the potential drop on a ballast resistor. Pulse voltage on discharge gap was measured with high-voltage probe with galvanic decoupling of input and output signals. Current and voltage pulses were registered using a digital oscilloscope.

Spectrograph ISP-30 equipped with photo-electronic cassette MORS-6 with a spectral range 196 - 930 nm was used for spectral diagnostics of the plasma. Exposure could be altered from 240 ms to 25 s. This provided a possibility of spectrum registration in linear regime of variation of the intensity of studied spectrum lines. The measurement was performed from 20 cm distance to the spectrograph slot: in the line of the hollow cathode axis and against the axis of plasma flame expansion.

Figure 1. Plasma source diagram.

Figure 2. Photos taken along the normal (a) and along the axis of propagation of the plasma flame (b).
3. Results and discussion

Figure 2a presents a photograph of the glow generated by plasma jet of the source when compressed air is used as a working gas. Maximum length of plasma jet as estimated by the zone of visible glow reaches 0.5-2 cm and varies depending on the features of the pulse discharge burning: frequency, amplitude, duration of discharge current pulses, the kind of the working gas and speed of its flow. Operation range of the average discharge current (pulse frequency) in which plasma jet is formed is 50-150 mA (2.5-7.5 kHz). The lower limit is connected with substantial weakening of plasma jet when the area of discharge working zone is decreased at the inner surface of a graphite cathode at low average currents (at low frequency). The upper limit of average current was restricted by maximum power of the used pulse power supply. The gas flow providing stability and cooling (nonequilibrium) of plasma jet varies in the range 1-10 l/min.

The photo of the sequence of radially distributing spark current channels (from the end of discharge system) is presented at figure 2b. The parameters of gas discharge system and gas flow are selected in such a way that chaotic movement of cathode spots of discharge inside the aperture in the graphite cathode generates a flow of secondary plasma distributed upon the total area of cathode outlet after the decay of numerous filamentary current channels. According to [4], low-current cathode spots on isotropic graphite are characterized by chaotic movement and lead to uniform erosion of all the working surface of the electrode.

The increase in the area of the inner surface of the cylindrical cathode at which cathode spots are generated and chaotically move, can be visually observed by the increase in the pulse frequency of the power supply (along with growth of average discharge current). When working at a minimum frequency of power supply ~10 Hz the spark channels are situated only at the shortest distance between the cathode and anode. The point of binding of current channels on the end of rod anode remains at that almost immovable from pulse to pulse.

In figure 3 the comparison of two oscillograms – of discharge current and voltage drop at ballast resistor when working with nitrogen is given for two magnitudes of pulse frequency of power supply – 70 Hz and 5 kHz. At high working frequency providing average discharge current at the level of 100 mA, weak current of glow discharge between pulses of magnitude 2-3 mA is observed. When the high voltage pulse is applied, spark breakdown of the gap happens with the monotonous current increase up to ~ 460 mA. At low frequency (70 Hz) the absence of current between pulses is characteristic and the growth of the main spark current takes place after passing of the pre-breakdown pulse with amplitude 18 mA with delay of the main pulse for 10 μs.

In figure 4 the waveforms of discharge current and voltage on the gap at average current 100 mA and frequency 5 kHz, working on nitrogen, are presented. The voltage of low-current glow discharge between pulses remains practically equal to discharge voltage (~ 500 V) during the main current pulse.

![Figure 3. Discharge current (channel 1) and voltage drop on 4 kΩ ballast resistor (channel 2) waveforms at nitrogen flow rate of 1.2 l/min for pulse frequency 70 Hz (a) and 5 kHz (b); time scale: 10 μs/div.; current scale: 150 mA/div.; voltage scale: 20 V/div.](image-url)
passing. The amplitude and duration of current pulses along with the change of the kind of gas and speed of flow are not substantially altered, being at the level of 500 mA and 40 microseconds (for values of average discharge current 100 mA and frequency 5 kHz).

Gas temperature in plasma jet was evaluated according to the data of spectral diagnostics basing on rotational temperature of \( \text{N}_2 \) molecules. Such estimate of temperature is allowed for nitrogen at atmospheric pressure because the condition of equilibrium between rotational and translational temperatures of molecules [5] is fulfilled. The rotational temperature was estimated by 337.13 nm band of the second positive system \( \text{C}_3\Pi_u \rightarrow \text{B}_3\Pi_g \) of nitrogen. The determined temperature of heavy plasma particles is \( 370 \pm 19 \) K at discharge current 150 mA. The temperature of the gas in the jet may be decreased to 50 °C along with the reduction of the average discharge current and increase in the distance from the cathode outlet.

The data of spectral diagnostics also permitted to evaluate the temperature of substance in cathode spots on the graphite cathode; its maximum value at the conditions of study is ~ 4000 °C. The process of carbon sublimation during the functioning of cathode spots at isotropic graphite allowed to get plasma containing a substantial part of CN-radicals as well as ions of atomic carbon, according to obtained optical emission spectra of plasma jet in nitrogen flow (figure 5). In addition to CN molecules, the highest peaks on the derived spectra of plasma jet were generated by \( \text{N}_2 \), \( \text{NO} \) and \( \text{N}_2^+ \) molecules. For plasma jet in air flow, beside radiation bands of excited molecules CN, \( \text{N}_2 \), NO and \( \text{N}_2^+ \), lines of atomic oxygen have sufficient intensity as well. The presence of these components in a plasma jet can be used for modification of properties and for plasma-chemical etching of the surfaces of materials,
including polymers because the developed plasma source permits selection of moderate temperature regime of treatment.

To study the effect of a plasma jet on carbon materials, a series of experiments on treatment of carbon fiber and pyrolytic graphite samples in air and nitrogen plasma was held. In figure 6 microphotos of carbon fiber treated in plasma made with the electronic microscope are shown.

![Figure 6. Microphotos of the surface of the carbon fiber after treatment in nitrogen (a) and air (b) plasma; (c) – the untreated carbon fiber surface.](image)

Because of plasma treatment of the samples, development of regular surface relief (figure 6) is observed that leads to the increase in roughness. The results of measurement of roughness parameter $R_a$, obtained with a probe microscope when scanning 1x1 $\mu$m$^2$ sites on samples of treated and untreated pyrolytic graphite, show approximately threefold increase in roughness after processing by air plasma of the studied source.

4. Conclusion

The characteristics of discharge and plasma of the atmospheric plasma jet source based on pulsed spark discharge with a hollow graphite cathode with coaxial geometry have been studied. Conditions of stable formation of plasma jet on the outlet of the source were determined for different plasma generating gases. It was shown that source plasma on base of air or nitrogen is non-equilibrium and temperature of heavy particles does not exceed $\sim$100 °C at average discharge current up to 150 mA.

In the composition of plasma jet in air or nitrogen flow, beside the main excited molecules specific for this class of devices, a substantial part of CN-radicals was observed. Spectroscopic temperature measurements on cathode surface indicate the presence of cathode spots on graphite having temperature sufficient for carbon sublimation. Functioning of such spots leads to saturation of plasma with CN molecules in nitrogen or air flow.

The hybrid composition of cold plasma jet including besides plasma forming gas, atoms and molecules formed during graphite sublimation allows to enhance the possibility of usage of cold atmospheric plasma sources for materials treatment and coating deposition.

References

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