Dielectric barrier discharge in radially converging gas flow generating two coaxial and opposite directed non-thermal plasma jets

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Abstract. An original source of non-equilibrium low-temperature plasma jets based on a barrier discharge in a radially converging flow of atomic and molecular gases at atmospheric pressure was developed and created. The discharge electrode system consists of two parallel and coaxial quartz disks, in the geometric center of those two coaxial identical holes were made. On the outer side of each disk a metal foil in the form of a wide ring is glued coaxially to the holes. The gas flow is directed from the periphery of the disks to their center and exits normally to the disks surfaces through narrow holes. As a result, two coaxial and opposite directed plasma jets are formed, which are perpendicular to the disks. There are no analogs of the developed source in literature. The source has been tested for plasma treatment of dielectric yarns that are transported through holes in dielectric barriers and are constantly enveloped in plasma jets. The results show practical possibility of using the developed gas-discharge source for continuous "roll-to-roll" plasma treatment of polymer filaments with the aim to improve their hydrophilicity.

1. Introduction
Non-equilibrium non-thermal plasma (NTP) is a source of various reactive particles (charged particles, electron, and vibrationally excited particles, radicals, UV-radiation) that can intensify or promote the needed processes. For this reason, NTP has been successfully used in various applications [1] for a long time, in particular, for surface treatment of thermally unstable materials in order to give them the necessary surface properties (improving wettability, increasing adhesion, etc.) [2-5]. Gas-discharge devices that generate NTP can be divided into two large classes-devices that work at low pressures of the plasma-forming gas (P < 1 Torr), and devices that work at atmospheric pressure. The main technical advantage of devices of the second class is the absence of expensive vacuum equipment, which also does not allow for a continuous technological process. There is also a difference in plasma chemical processes that are dominant in low-pressure and atmospheric NTP. Thus, in the processing of materials with NTP created by a low-pressure RF discharge, the main role is played by positive ions that bombard the treated surface with the energy of several tens of eV. This energy is acquired by ions in the layer of spatial charge formed near the treated surface in contact with the plasma. In the case of atmospheric pressure NTP, the main role in bulk and surface plasmochemical processes is played by electron and vibrationally excited particles and radicals [6].

To date, a wide variety of gas-discharge devices have been created that generate NTP in atomic and molecular gases at atmospheric pressure. In some devices, plasma processing is carried out in the discharge zone, while other devices create plasma jets that perform remote processing of materials and
objects. In the latter case, a very important task is to create a high concentration of electron and vibrationally excited particles and radicals in the plasma jet.

To solve this problem, we have developed a plasma source based on a barrier discharge with a radially converging flow of atomic gases, which forms two coaxial plasma jets pointed in different directions. A unique feature of the new plasma source is that the discharge that creates active particles is formed between the disk electrodes over a large area determined by the effective area of the barriers. At the same time, the radially converging gas flow transfers all long-lived active particles to the central exit holes, whose cross-section is much smaller than the area occupied by the discharge. As a result of forced geometric focusing (cumulation) of the flow, the concentration of active particles in the plasma jets coming out of the holes is much higher than their average concentration in the discharge zone. The article presents the results on the measurements of the electrical parameters of the new source of plasma jets and a successful example of its use to improve the wettability of cord yarn used for reinforcement of automobile tires.

2. Experimental setup
The general scheme of the barrier discharge in a radially converging flow either atomic or molecular gases at atmospheric pressure is shown in Fig. 1a. The barrier discharge electrode system consists of two quartz disks disposed of in parallel and coaxially to each other. In the geometric center of both disks, there are coaxial identical holes with a diameter of 2 mm. On the outer side of each disk, a metal foil in the form of a wide ring is glued coaxially to the holes. The gas flow enters between disks and is directed from the periphery of the disks to their center and exits normally to the disks surfaces through narrow central holes. Once the barrier discharge is formed between the dielectric disks, plasma is transported by the gas flow to the center and, at the outlet, two coaxial plasma jets are formed, perpendicular to the disks and directed in the opposite directions. The appearance of a laboratory-scale installation for continuous "roll-to-roll" processing of polymer yarns and filaments in coaxial and oppositely directed plasma jets is shown in Fig. 1b. The yarn to be processed with a diameter of about 1 mm is continuously transported through the holes made in the dielectric barriers so that it is constantly enveloped by plasma jets along their entire length. As far as the authors know, in the literature, there are no analogs of this developed source forming simultaneously two coaxial and opposite directed plasma jets.
Figure 1. a) The electric scheme of the barrier discharge in radially-converging gas flow includes the following components: the quartz disks ($\varepsilon \approx 4$) of 5 mm in thickness with the holes of 2 mm in diameter for gas and plasma efflux, the metallic foil in a form of ring with the outer diameter of 40 mm, the polymer yarn to be processed of 1 mm in thickness. b) The photograph of the laboratory scale device for “roll-to-roll” processing of a dielectric yarn inside the opposite directed plasma jets formed by the barrier discharge in radially-converging gas flow. Two upper insets show the enlarged images of yarn enveloped by He plasma jets.

3. Experimental results.

At first, we conducted some experiments to excite a barrier discharge in a radially converging airflow at atmospheric pressure powered with the sinusoidal voltage frequency of 50 Hz and 85 kHz. It turned out that, despite the fact that the discharge in the airflow was ignited, there were no visible plasma jets in the airflows escaping the source. A possible reason is the insufficient barrier discharge power, which was limited by both the air electronegativity and the dielectric barriers with an excessively large thickness (5 mm). Indeed, the maximum discharge power in the airflow was only $P = 0.2$ W at the voltage frequency of 50 Hz and a voltage amplitude of $U = 22.4$ kV and $P = 0.2$ W at the voltage frequency of 85 kHz and the amplitude of $U = 6.8$ kV. For this reason, the main studies of barrier discharge were conducted in radially converging streams of helium and / or argon at a sinusoidal voltage frequency of 85 kHz with an amplitude $U$ up to 7 kV.

During the experimental investigation of the developed gas-discharge system electrical characteristics, it was found that at the fixed amplitude of the applied voltage, such parameters as the discharge current and discharge power change weakly when the dielectric yarn is placed in the plasma jets (see Fig.2). This circumstance makes it possible to use a barrier discharge in a radially converging flow for plasma treatment of non-conducting filamentous objects whose diameter does not exceed 1 mm.
Figure 2. Voltage and current waveforms of the barrier discharge in radially-converging helium flow at atmospheric pressure. Gas flow rate through the openings to the atmosphere is 40 m/sec. a) Voltage and current waveforms when there is no yarn in the discharge, the power is 8.6 W; b) Voltage and current waveforms when the yarn is being processed, the discharge power is 10.5 W.

Figure 2 demonstrates some increase in the discharge current and power (by about 20%), which occurs in the presence of a yarn. A reason for that may be due to the partial locking of the output holes with the yarn. In this case, there is an increase in the gas residence time in the discharge zone that leads to additional gas heating. Because of the gas heating, the gas density $N$ diminishes, the reduced electric field $E/N$ magnitude increases that results in the intensification of ionization processes and, accordingly, leads to a certain increase in the discharge current and the discharge power. Generally speaking, this is a positive effect, which in combination with the increase in the gas residence time in the discharge zone, leads to the increase in the number of reactive particles created by the discharge at a given amplitude of the applied voltage.

4. The results on plasma treatment and discussion.
A few explanatory words about why a given polymer yarn was chosen to test the effectiveness of the new plasma source, specifically, cord yarn used for reinforcing car tires. Increasing the grip strength of the cord yarn with the rubber of a car tire is one of the key factors that affect the consumer quality of the tire. In turn, the adhesion of the yarn to the rubber depends on its hydrophilicity [7]. Currently, several plasma methods used for hydrophilization of polymer filaments, in particular, cord filaments, are known. One of them uses a low-pressure NTP source based on an RF discharge with a power of about 1.5 kW [8]. A positive effect of NTP processing has been demonstrated, but the required NTP processing times correspond to several minutes (up to 10 minutes). In [9], yarn processing in NTP was performed in the zone of a plane-to-plane barrier discharge at atmospheric pressure. It is shown that within the frequency range of the supply voltage of 50 Hz - 10 kHz, the effect of plasma processing is practically absent, but occurs in the region of higher frequencies. In [10], the yarn was also processed at atmospheric pressure, but with the usage of two independent jet plasma sources, the jets of which were directed at an angle to each other, and in the area of their intersection, the processed yarn was placed. In this case, the effective length of the processing area was less than the length of each of the jets. The power of the sources was very high (about 1.5 kW each), therefore the plasma jets were hot. The power of the sources was very high (about 1.5 kW each), therefore the plasma jets were hot. Due to that, there was a problem with the thermal elongation and sagging of the yarn, which led to the problem of collinearity of the yarn and plasma area and respectively to inhomogeneity of the yarn processing.

Thus, the experiments performed on NTP processing of polymer filaments showed the prospects of the plasma method, but at the same time, found problems related to the need of reduction of the processing time and power of the plasma sources used. Taking this into account, the low-power gas-
discharge source, which is able to generate simultaneously two coaxial and opposite directed plasma jets, is very promising for NTP processing of polymer filaments. The relevant experiments were carried out and the results are presented below.

By now, quantitative characterization of hydrophilicity of the treated objects is carried out by different methods, depending on the shape and structure of objects. We will point out the simplest and most accessible methods. Thus, in the case of flat objects with a solid surface, goniometry is used to determine their wettability [11,12], i.e. the value of the contact angle of the liquid drop with the surface before and after plasma treatment is measured. In the case of a fiber or porous structure of the object (such as tissues and similar objects), the height of the liquid rise along the vertical fabric, the lower edge of which is lowered into the liquid [13], is measured. However, in the case of a yarn-like object, these methods are not applicable. The first method is not applicable for geometric reasons, since a thin yarn is almost a one-dimensional object. The second method is not applicable because the small cross-section of the yarn strongly restricts the rate of liquid rise. In this case, the height of the liquid rise is determined not only by the wettability of the yarn, but also by the rate of evaporation of the liquid during its rise, which greatly complicates the interpretation of the results.

To evaluate the effectiveness of the polymer yarns plasma treatment aimed to improve their hydrophilicity, we developed a simple and original method that has no analogs in the literature. The method is based on taking into account the fact that the cord yarn to be processed is not actually solid, but is woven from a large number of very thin fibers. The fibrous structure forms the potential hygroscopicity of the yarn. The first step of this method consists of deposition with a special syringe of a very small droplet of distilled water with a volume of 5 microliters on a horizontally positioned multi-fiber yarn. Then the complete absorption time of the droplet into the yarn was recorded. The set of images shown in Figure 3 explains the idea of the method.

![Figure 3](image_url)

**Figure 3.** A series of photographs showing the dynamics of the absorption of a distilled water drop deposited on a multi-fiber polymer yarn. The photographs indicate the time since the deposition of the water drop on the yarns. Photos of the upper row correspond to the yarn treated by plasma for 1 minute; bottom row photographs correspond to the untreated yarn. If compare the sizes of initial water droplets (t = 0 s), there is a false impression that the initial droplet deposited on the treated yarn is smaller than that deposited on the untreated yarn. In fact, the initial volume of the droplets is the same. A reason is that the treated yarn very quickly absorbs some portion of the deposited droplet during the deposition process (about of 0.5 s).

Verification by the proposed method of the hydrophilicity of the cord yarn processed in the "roll-to-roll" mode in plasma jets of the barrier discharge in radially converging helium flow showed that
the yarn hydrophilicity actually increases. This is manifested in a significant (more than twice) acceleration of the absorption of the water drop into the cord yarn after its plasma treatment (see Fig. 3). Besides, we have done the additional test. After the vulcanization of rubber with treated and untreated yarns, mechanical tests were performed on the determination of the strength of the yarns adhesion to the rubber. It turned out that to pull the plasma-treated yarn out of the rubber, it is required to use the mechanical force that is about 50% higher than the force used for the untreated yarn. Thus, the performed research proved the prospects and high efficiency of the developed gas discharge source of two coaxial and opposite directed non-thermal plasma jets when it is used for plasma hydrophilization of filamentous objects in a continuous “roll-to-roll” mode.

5. Conclusions
Plasma treatment of polymer filaments at atmospheric pressure and in continuous mode requires the development of specialized gas-discharge sources. The article describes the original source of non-equilibrium low-temperature plasma jets created by a barrier discharge in a radially converging flow of atomic and molecular gases at atmospheric pressure. The gas flow is directed from the periphery of parallel and coaxial disk barrier electrodes to their center. Then it goes out normally to the surface of the barriers through narrow holes, forming simultaneously two coaxial and multidirectional plasma jets. There are no analogues of the developed plasma jets source in the literature.

A unique feature of the new plasma source is that the barrier discharge that creates active particles is formed between two dielectric disks over their large area. At the same time, all long-lived reactive particles formed by the discharge are transferred by the radially converging gas flow to the central exit holes, whose cross-section is much smaller than the area occupied by the discharge. As a result, there is a forced cumulation of the reactive particles at the center due to geometric focusing of the flow. Because of that, the concentration of reactive particles in the plasma jets coming out of the holes is much higher than their average concentration in the discharge zone, especially at the periphery.

A low-power two-jet source was tested applying the plasma to the cord yarn used for reinforcing automobile tires. During NTP processing, the yarn is transferred through holes in the dielectric barriers and is constantly enveloped by non-equilibrium plasma jets. The presence of two jets increases the contact time of the yarn to be treated with the active plasma. The results of the work show the prospects and practical possibility of the usage of the created gas-discharge source for continuous "roll-to-roll" plasma processing of dielectric polymer filaments with the aim to improve their hydrophilicity and adhesive properties.

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