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Peculiarities of the formation of a filamentary structure of a microwave discharge in an argon flow

© S.V. Sintsov, A.V. Vodopyanov, A.N. Stepanov, D.A. Mansfeld, N.V. Chekmarev, E.I. Preobrazhensky, A.A. Murzanev, A.V. Romashkin

Institute of Applied Physics, Russian Academy of Sciences, Nizhny Novgorod, Russia
e-mail: sins@ipfran.ru

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This paper presents the results of an experimental study of the spatial structure of a microwave discharge maintained in an argon flow by gyrotron radiation in a continuous mode with a frequency of 24 GHz at atmospheric pressure. In the structure of the plasma plume, stationary filamentary channels are observed, elongated along the direction of the argon flow, regardless of the orientation of the external electric field of the wave, surrounded by a diffusion halo. Measurements of the electron density, vibrational and rotational temperatures of gas molecules in plasma filaments have been carried out. The role of gas-dynamic mechanisms responsible for the formation of the inhomogeneous static structure of the plasma torch and the maintenance of a substantially nonequilibrium distribution of temperature characteristics in the discharge is discussed.

Keywords: high-pressure microwave discharge, plasma torch, argon, filamentous plasma channels, filaments.

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Introduction

Studying of nonequilibrium high pressure microwave discharges is an important applied task. The relevance of research activities in this field of the plasma physics is attributable first of all to potential industrial applications in the problems of nonequilibrium plasma chemistry [1–14]. Microwave discharges have highly localized energy in the region of support, limited by the geometry of microwave beam or a waveguiding structure with a characteristic spatial scale of an order of wavelength of the heating field. This is responsible for the high power density in the region of discharge support and the possibility of its removal from chamber walls, which is important for running high-temperature processes and working with chemically reactive compounds [2,4,8]. Microwave discharges of wide pressure range are characterized by high electron density close to the critical value for the heating field frequency, which is also important from the point of view of efficiency improvement and increase in the speed of plasma-chemical synthesis processes [10–14].

High pressure microwave discharges have a heterogeneous spatial structure [15–25]. Often it is defined by the presence of thin filament channels in the plasma volume predominantly oriented along the electric field of the wave. The mechanism of emergence of such filaments is considered to be related to the development of ionization-overheating instabilities (IOS) on fluctuations of plasma parameters in the discharge [15–25].

Previously, our research group has conducted a study of the stationary nonequilibrium plasma torch supported by continuous millimeter-band radiation in an argon jet penetrating into the ambient air at atmospheric pressure [10,26,27]. The discharge was supported in an argon flow in the waist region of a microwave beam with a frequency of 24 GHz and a power of up to 5 kW in the continuous mode. It was shown that temperature of electrons in the plasma torch is 5−7 times higher than the temperature of gas. This work presents results of the experimental study of spatial structure of such subthreshold microwave discharge. It turned out that stationary filament plasma channels are observed in this discharge, which are stretched along the direction of the argon flow, gas temperature in which is considerably higher than the mean temperature for the plasma torch. Orientation of these filament channels is independent on the direction of electric field of the wave, which haven't been observed in microwave discharges before [4,9,15–20]. In this work we discuss the role of gas-dynamic mechanisms responsible for formation of the stationary filament structure of plasma torch and support of a considerably nonequilibrium distribution of temperature characteristics in the discharge.

1. Experimental setup

The experimental setup is shown in Fig. 1. A 24 GHz process gyrotron with a power of up to 5 kW in continuous mode was used as a source of microwave radiation[26,27]. The output Gaussian beam with a linear polarization was entered into a cylinder gas-discharge chamber, where it was focused by a parabolic mirror at a distance of 32 cm from it along the chamber axis. Length of beam waist is 11 cm, maximum power density in this region is up to 3.4 kW/cm^2, root-mean-square strength of the field is 1.1 kV/cm [26,27].

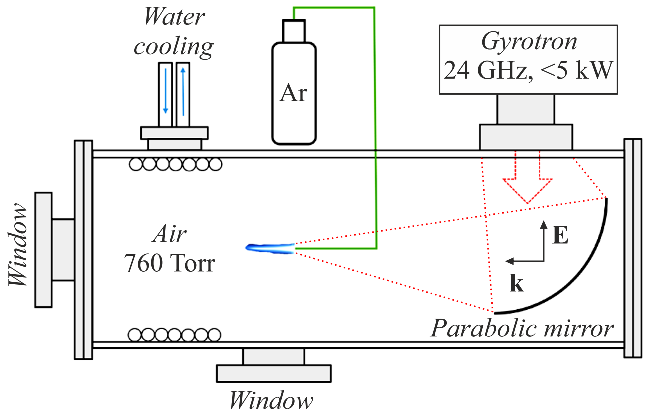

Figure 1. Scheme of the experimental setup.

Along the symmetric axis of the beam, in the region of its waist, a metal tube was placed with an outer diameter of 4 mm and an inner diameter of 2 mm to feed argon. Position of the gas inflow tube cut corresponded to the center of the waist region of quasi-optic beam. The argon flowrate was varied in the range from 3 to 50 l/min. The continuous discharge was initiated using high-voltage spark discharge in an argon jet flowing into the air at atmospheric pressure. The emerging plasma torch was „linked" to the gas inflow tube cut and stretched along the direction of argon flow parallel to the wave vector of the microwave beam. Diameter of the plasma torch is equal to the outed diameter of the metal gas inflow tube. Cross-section area of the plasma torch was about 10 times less than the cross-section area of the beam in the waist region.

On the side surface of the cylinder gas-discharge chamber, approximately at the waist region of the microwave beam, a window was located through which the discharge diagnostics was carried out. Also, a calibrated water-cooled calorimeter was accommodated inside the chamber, that allowed measurement the portion of power not absorbed by the discharge. Power absorbed by the discharge was calculated as the difference between the output power of the microwave source and the power absorbed by the calibrated calorimeter. For all modes of support, the power absorbed by the discharge was about 10 times less than the microwave power introduced into the gas-discharge chamber, which approximately corresponded to the ratio of cross-section areas of the plasma torch and the quasi-optical beam in the waist region.

2. Plasma torch structure

Fig. 2 shows photo of a microwave discharge at atmospheric pressure supported by continuous radiation at a frequency of 24 GHz. We have been succeeded to initiate the discharge with the minimum output power of microwave radiation at a level of 0.9 kW, which corresponded to a root-mean-square electric field of about 0.6 kV/cm in the focus. An increase in heating power up to 5 kW resulted in linear increase in the luminous plasma region up to 42 mm without change in its structure and diameter. A change in the argon flow in the range from 5 to 20 l/min had no effect on shape and size of the discharge [26]. The plasma torch was composed of filament channels and a diffusion halo around them (Fig. 2). Filaments were stretched along the direction of gas flow and „linked" to the gas inflow tube cut. With the minimum power of discharge support 1 filament was observed in the torch, and with the maximum power their number increased up to 10–12. With increase in the heating power the length of filaments increased together with the length of the plasma torch itself, while their diameter remained unchanged. The position of plasma filaments was found to be fixed at the gas inflow tube cut. Its rotation about its axis resulted in appropriate shift of each filament position. This is indicative of the fact that the place of plasma filaments emergence on the cut of metal gas inflow tube is most likely to be attributable to the presence of irregularities with a characteristic size not more than 100 μm.

Indeed, at the moment of discharge initiation with the minimum heating power of 0.9 kW, the electric field in the region of discharge is about 3 times less than the breakdown level for argon at normal conditions [28]. A local amplification of the field by several times at the cutting edge resulted in fulfilment of the condition of existence of a self-sustained discharge support region. With increase in the external microwave electric field the breakdown conditions starts to be fulfilled on other irregularities of the gas inflow tube cut as well, which results in increase in regions of self-sustained discharge existence and, as a consequence, increase in the number of plasma filaments.

To verify this hypothesis, special metal tube-nozzles were manufactured with an outer diameter of 6 mm and an inner diameter of 4 mm, that were put on the initial gas inflow tube (Fig. 3, a). 6 triangular prongs with a characteristic size of 0.7 mm were made at the tube-nozzle

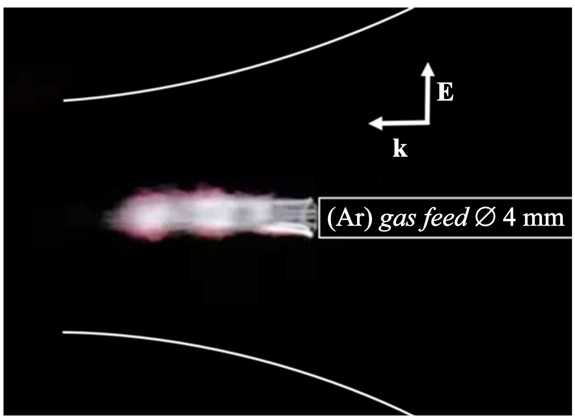

Figure 2. Photo of a plasma torch supported in a flow of argon by continuous microwave radiation at a frequency of 24 GHz in the air at atmospheric pressure. Argon flowrate is 15 l/min, microwave radiation power is 4 kW.

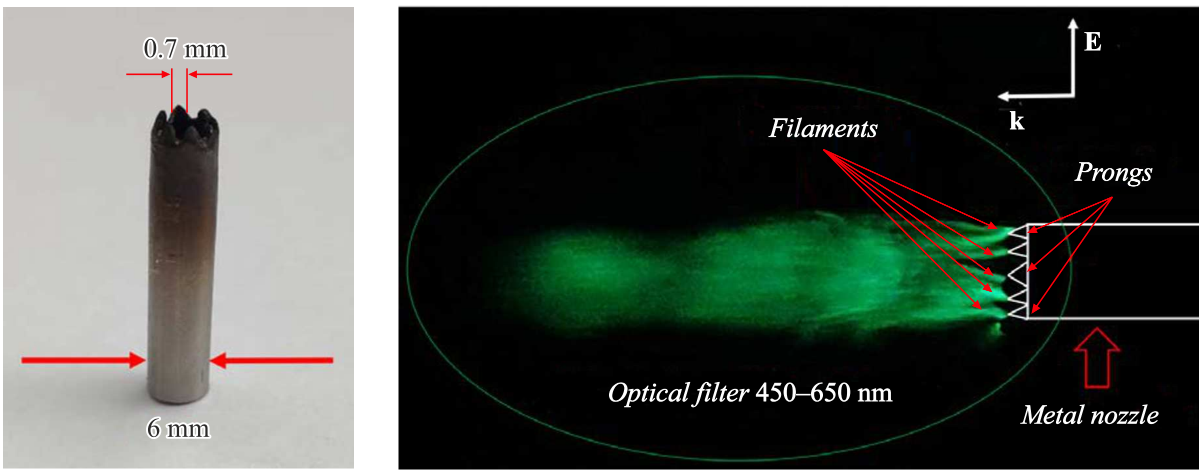

Figure 3. *a* — photo of the tube-nozzle with six prongs; *b* — photo of the plasma torch supported at the cut of the tube-nozzle.

cut. Fig. 3, *b* shows photo of a plasma torch initiated in an argon flow at the cut of the tube-nozzle with six prongs. The photo was snapped through an optical filter with a bandwidth of 450−650 nm for the best demonstration of the plasma filaments contour. With a heating power over 2 kW filaments were observed in the plasma torch, which number was equal to the number of prongs on the metal tube-nozzle.

Thus, the emergence of filaments was unambiguously linked to each of the prongs existing on the tube-nozzle. Further increase in the heating power resulted in emergence of new filaments. It is worth noting that in contrast to the experiment with an even gas inflow tube cut (size of irregularities is less than 100 μm), filaments have shorter lengths and become slightly distorted in relation to the main direction of the argon flow. This is connected to the fact that the flow of plasma-generating gas (argon) is distorted while flowing over the metal prongs. In addition, together with the gas flow on irregularities of the gas inflow tube cut the orientation of the electric field becomes distorted as well, which also could contribute to the filament distortion. To find out the role of orientation of the microwave electric field in the formation of filaments, an additional series of experiment was conducted.

The orientation of the gas inflow tube cut (without the nozzle, with an outer diameter of 4 mm) was changed in relation to the direction of wave propagation and direction of the electric field strength vector. Fig. 4 shows photos of discharge at different orientations of wave vector and electric field of the wave in relation to the direction of plasma-generating gas flow. Fig. 4, *a* — initial orientation investigated in previous works, where argon flow was oriented along the direction of microwave field propagation and normal to the electric field. Fig. 4, *b* — the argon flow was fed at an angle of 40° in relation to the direction of wave propagation and at an angle of 50° in relation to the electric field of the wave. Fig. 4, *c* — direction of the gas flow normal to both the wave vector and the electric field vector (photo is snapped through the window in the end of the chamber). Fig. 4, *d* — direction of the gas flow parallel to the electric field strength vector and normal to the wave vector. In all cases spatial position of the gas inflow tube cut was selected such that the plasma torch was completely within the beam waist region. In can be seen, that bright filament channels emerge in the plasma torch independent on the orientation of gas flow direction in relation to the microwave beam. At any orientation of the gas inflow tube, filaments emerged at its cut and were stretched along the gas flow direction. This is indicative of the fact that the mechanism of heterogenous stationary discharge structure formation in the gas flow was defined mainly by the running gas-dynamic processes.

3. Optical interferometry of the plasma torch

Using the laser optical interferometry the spatial distribution of refractive index of gas in the region of plasma torch support was investigated. The essence of this method consists in determining of the additional phase shift of the sounding laser beam, which is resulted from the passage through the region containing the plasma torch. Since the plasma concentration in the discharge is $\sim 10^{16}\,\mathrm{cm}^{-3}$, the contribution of the electron component to the refractive index change is much less than its change related to the decrease in the gas density due to heating, then the interferometric data allows obtaining an isobaric approximation of the gas temperature distribution in the region of discharge support [29].

In the experiment we used the Michelson interferometer scheme. We used a continuous semiconductor laser with a

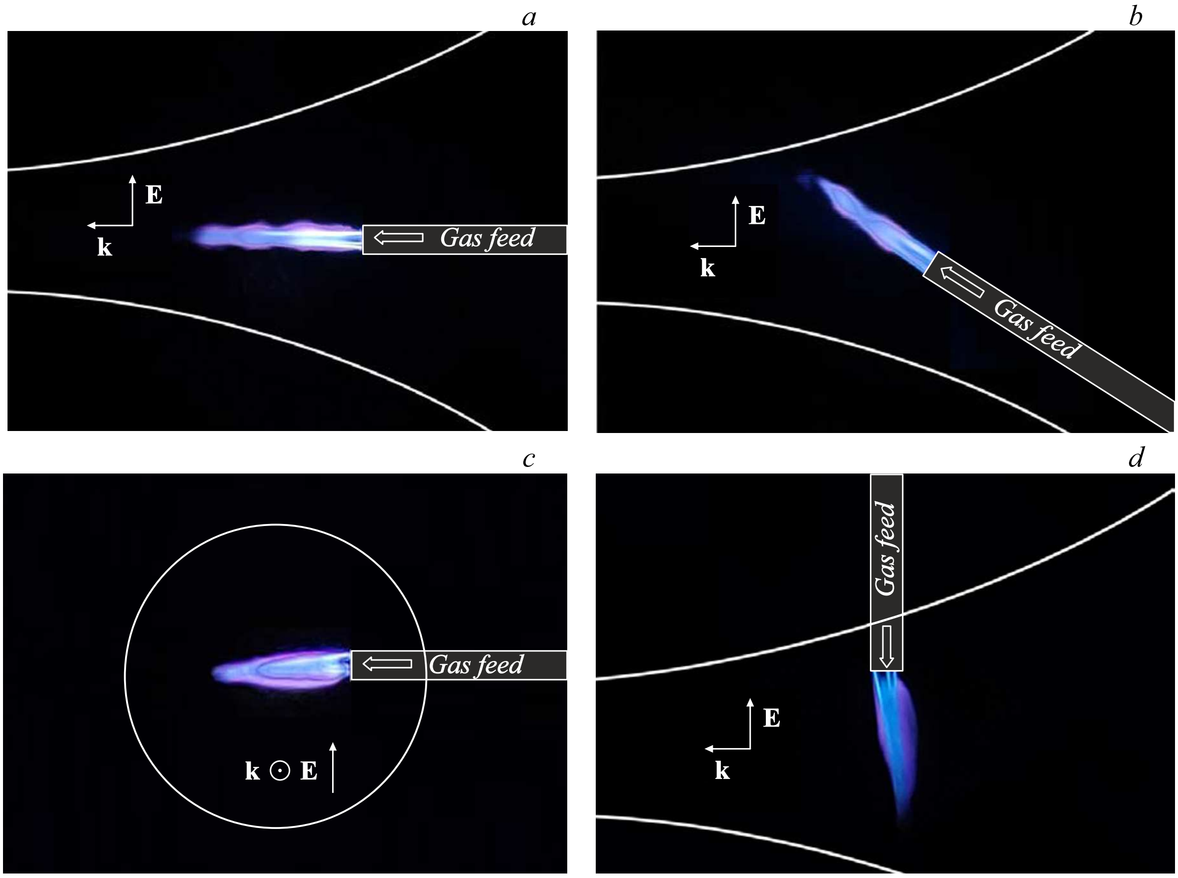

Figure 4. Photo of the plasma torch at different orientations of the wave vector and the electric field of the wave in relation to the direction of argon flow.

wavelength of 532 nm and a power of 10 mW as a source of the sounding radiation. The sounding laser beam of a larger diameter passed through the region where plasma emerged. The part of the beam, that was overpassing the discharge served as a reference signal in the interferometer scheme.

For the numerical processing of obtained interferometer patterns we used IDEA 1.7 software package[30]. Fig. 5 shows the image of sounding radiation phase shift distribution in the region of discharge support, exposure time is 3 s, microwave radiation power is 0.9 kW. The blue rectangle on the left shows position of the gas inflow tube cut. The sole-color blue region in the top right corner shows the limit of visibility of the sounding beam.

The spatial distribution of phase caused by the single filament emerging in the discharge in this mode can be clearly seen in the figure. With a higher heating power, when there are several filaments in the torch, the distribution image becomes more difficult for interpreting because of superposition of such phase distributions from several plasma filaments.

The gas temperature was estimated on the basis of the obtained pattern of sounding radiation phase distribution using Abelian transform in an assumption of cylindrical symmetry of the plasma torch. Sensitivity of the conducted interferometric measurements did not allow us to determine the gas density, that corresponds a temperature over 1400 K. As a result of the estimates made, it was fount that

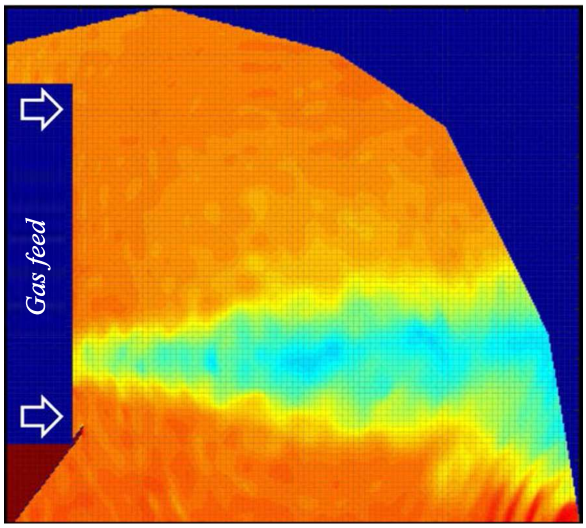

Figure 5. Interference image of sounding radiation phase shift distribution in the region of discharge support.

gas temperature within the cone contour of a filament is not more than 1400 K, and in the surrounding region of plasma halo it is not more than 600 K. Color palette of the image within the cone region of the most distorted phase shift remains unchanged, since it is in saturation in terms of sensitivity. Inside the saturation region there is a more high-temperature filament region, corresponding to the contour of visually observe filament in the optical range. It can be seen in the obtained pattern of the sounding radiation phase distribution how the hot gas from the filament expands with distance from the tube cut and forms a high-temperature zone of discharge at a distance of 4–5 mm from the gas inflow tube cut. An assumption can be made that the expansion of excited gas particles and free diffusion of charge carriers from filaments improve the breakdown conditions in the halo around them, which is a region of nonself-sustained discharge. The emission of filaments in ultraviolent (UV) range of wavelengths and the mechanism of gas-dynamic carry-over of plasma from the high-temperature zone of plasma channels may define a significantly nonequilibrium distribution of temperature characteristics implemented in the plasma halo in accordance with previous measurements [10,26,27].

4. Optical emission spectroscopy of the plasma torch

With an argon gas flowrate from the gas inflow tube more than 30 l/min a specific mode of discharge support was implemented, where we have succeeded to separate plasma filaments from the plasma halo. Fig. 6 shows photo of a plasma torch in the mode with a high rate of argon feed. It can be seen that in the region near the gas inflow tube cut „bare" filaments are observed. At a distance of 6 mm from the gas inflow tube cut the plasma filaments are surrounded by a halo, as they were at lower argon flowrates (Fig. 2).

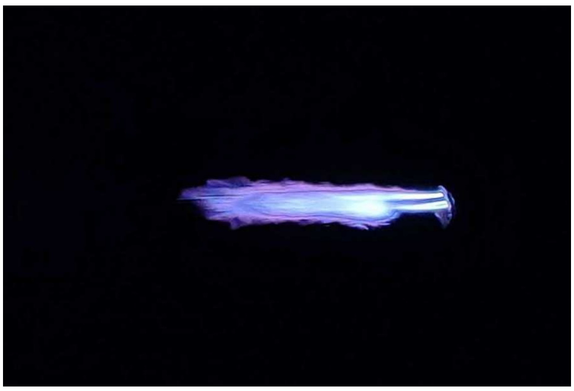

Figure 6. Photo of a plasma torch supported in a flow of argon by continuous radiation at a frequency of 24 GHz in the air at atmospheric pressure. Argon flowrate is 40 l/min, microwave radiation power is 3 kW.

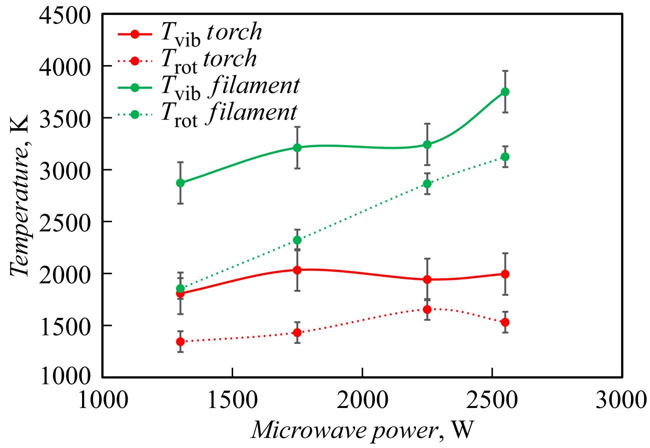

Figure 7. Dependencies of vibrational and rotational temperatures of nitrogen in the plasma torch (red (in on-line version)) and in filaments (green (in on-line version)). T_{vib} — vibrational temperature; T_{rot} — rotational temperature.

Emission spectra were obtained using a MS5204i spectrometer-monochromator with Czerny-Turner configuration by SOL Instruments with highly-sensitive CCD-array. Spectra were recorded in the range of 300–000 nm with a sampling rate of 0.028 nm. Width of the spectrometer hardware function was determined from the radiation of a calibration neon lamp and was equal to 0.15 nm [31,32]. Emission spectra were recorded through the quartz window located on the side of the gas-discharge chamber, opposite to the region of microwave beam focusing (Fig. 1). Using a system of quartz lenses a torch image was formed, that allowed obtaining separately the emission spectra for the region in front of the gas inflow tube cut with „bare" filaments and the remote region of discharge, i.e. the plasma torch, which is composed of filaments and a diffusion halo around them. Based on electron-vibrational-rotational spectra of the second positive system of nitrogen mixed in the plasma torch from the ambient air atmosphere, vibrational and rotational temperatures in filaments and in the plasma torch were determined. The estimation technique widely known and described in detail in [33–36]. Due to the high velocity of collisions at the atmospheric pressure in the conditions of experiment, the rotational temperature is approximately equal to the gas temperature, which will be used in the following for estimates.

Fig. 7 shows results of the measurement of above-mentioned temperature characteristics. It can be seen that vibrational and rotational temperatures of nitrogen in filaments at 1000–1500 K is higher than respective temperatures in the plasma torch. The rotational temperature of nitrogen molecules increases with growth of the input power from 2000 to 3000 K.

At a gas temperature level of 3000 K the effective gas pressure (pressure of the environmental gas multiplied by the temperature ratio between the environment and the plasma) in filaments is equal to approximately 80 Torr, which is close to the minimum of the breakdown curve of argon for electromagnetic radiation with a frequency of 24 GHz and a diffusion length of 0.4 mm [28]. With such effective pressure the external microwave electric field has breakdown level of field strength, and discharge in the filaments is self-sustained. Temperature characteristics of the plasma torch itself, which is composed of filaments and a halo, corresponds approximately to the mean temperatures of filaments and plasma halo measured in previous works [26,27]. It can be seen that an increase in the heating power, that results in a torch length increase, does not entails an increase in integral temperature characteristics of the plasma torch, which is indicative of a large number of inelastic collision processes with molecules.

On the other hand, vibrational and rotational temperatures of nitrogen in filaments grow noticeably with increasing power of heating, because the portion of mix in molecular gases in them is considerably lower.

The electron density was measured by the broadening of H_α and H_β hydrogen lines of Balmer series due to the linear Stark effect. Hydrogen was initially mixed in the plasma-generating gas, its concentration was not more than 5%. The technique of electron density counting by this method is described in detail in [37–41]. As a result, it was found that broadening of the above-mentioned lines in the plasma torch does not exceed a value close to the width of the spectrometer hardware function. Taking into account available accuracy of measurements of emission spectra, the broadening of hydrogen lines in the torch corresponds to an electron density less than $10^{14}\,\mathrm{cm}^{-3}$. The broadening of Balmer series hydrogen lines in filaments is noticeably higher than the width of the spectrometer hardware function and the broadening caused by Doppler and Van der Waals mechanisms. Recalculation of the hydrogen lines broadening in filaments caused by the linear Stark effect allows estimating the electron density at a level of $(7 \pm 3) \cdot 10^{14}\,\mathrm{cm}^{-3}$. The electron density within the confidence interval was independent on the heating power (with argon flowrate fixed and higher than 30 l/min). The obtained estimate of electron density in filaments is two orders of magnitude higher than the critical value for the external field frequency, which is $7 \cdot 10^{12}\,\mathrm{cm}^{-3}$. It is worth noting that the rotational temperature in the plasma filaments is measured by the emission radiation of nitrogen, which is mixed in from the ambient atmosphere. Therefore, the gas temperature estimates obtained by this method mostly characterize temperature of the outer shell of bare filaments. The electron density measured by broadening of hydrogen emission lines contains information on the internal regions of filaments, where temperature may be higher. Filaments are a region of self-sustained discharge, therefore temperature characteristics inside filaments have equilibrium distribution, and the electron density is directly related to the gas temperature by the Saha equation [28]. Then the measured electron density inside filaments corresponds to a gas temperature approximately equal to 7000 K. However, we did not manage to confirm this estimate experimentally.

5. Discussion

Results of the experiments performed are indicative of the fact that structure of the continuous microwave discharge in the argon flow at atmospheric pressure is considerably heterogeneous. Filament plasma channels are observed, being oriented along the direction of argon flow regardless of the direction of external electric field of wave. It means that the mechanism of formation of such filaments is directly caused by gas-dynamic stationary processes running in the region of discharge support. At the moment of discharge initiation in a gas flow the conditions of breakdown are only fulfilled in the region of field amplification on irregularities of the gas inflow tube cut, which results in formation of a strongly localized region of self-sustained discharge, „blown over " by cold plasma-generating gas. It is this point that „links" the emerging stationary filaments to the gas inflow tube cut. Further pulling out of filaments from the localized region of self-sustained discharge can not be caused by the development of ionization-overheating instabilities, because in pre-breakdown conditions of discharge support the growth of ionization frequency on fluctuations of plasma parameters is limited. Then the pulling out of plasma channels may be related to thermal-conductivity heating of the gas at the leading front of the point area of self-sustained discharge. The direction of filaments pulling out is defined by the direction of gas flow, and their lateral growth is limited by the flow of „cold" plasma-generating gas that is blowing over the filaments. Gradual thermal-conductivity heating in the forming plasma channel decreases the gas density until the external electric field of the wave becomes the breakdown field, and discharge becomes self-sustained in continuous mode.

This mechanism of stationary filaments formation in a subthreshold gas flow matches well the obtained experimental estimates of plasma parameters. It was reliably demonstrated that gas temperature in the filaments is considerably higher than gas temperature in the plasma halo and is equal to $2000-3000\,\mathrm{K}$, depending on the heating power. With a gas temperature at a level of 3000 K, the effective gas pressure in filaments is equal approximately to 80 Torr, which is close to the minimum of breakdown curve of argon for electromagnetic radiation with a frequency of 24 GHz. With a temperature of 2000 K, the effective pressure is equal approximately to 120 Torr, and electric field strength of the wave in the region of discharge support achieves the breakdown level [28].

The joint evolution of electromagnetic field and plasma in conditions of continuous microwave discharge in a gas flow results in effective heating of stationary plasma channels. It was shown experimentally that electron density in the filament channels is 2 orders of magnitude higher than the critical level for the frequency of heating field and is equal to $(7 \pm 3) \cdot 10^{14}\,\mathrm{cm}^{-3}$. This is indicative of possible transformation of microwave field to surface wave that provides for self-consistent heating of supercritical plasma along all the length of filament channels. The estimates performed show that the length of propagation of such surface wave directed by filaments may be much greater than the diameter of plasma channel and the depth of skin-layer, and limited by the size of the strong field region in the waist region of the microwave beam. On the other hand, filaments are formed independently from the orientation of electric field of the wave, therefore, at this stage of research, it is impossible to state unambiguously that a mechanism of microwave radiation transformation to surface wave exists.

UV radiation of filaments and gas-dynamic carry-over of excited particles from the filament region of self-sustained discharge results in the effective absorption of microwave field in the region of nonself-sustained discharge, the plasma halo, which is visually amount to the main volume of the plasma torch. By all appearances, such nonequilibrium mechanism of gas excitation in the region of nonself-sustained discharge allows a significantly nonequilibrium distribution of temperature characteristics to be realized in the plasma halo, that were measured in previous works [26,27]. This type of nonequilibrium continuous discharge of atmospheric pressure can be used to solve a number of problems of the modern industrial plasma-chemistry [1–14]. The efficiency of its use to destruct molecular compounds with high bonding energy is demonstrated by means of an example of the carbon dioxide utilization task [10].

Conclusions

As a result of the performed experimental study of continuous microwave discharge in an argon flow at atmospheric pressure it was shown, that formed filament plasma channels are oriented along the direction of plasma-generating gas flow regardless of the direction of external electric field of the wave. It means that the mechanism of formation of such filaments is directly caused by gas-dynamic stationary processes running in the region of discharge support. Temperature characteristics of plasma channels are considerably higher than corresponding mean values for the plasma torch, and the electron density is 2 orders of magnitude higher than the critical level for the heating field frequency. UV radiation of filaments and gas-dynamic carry-over of excited particles from the filament region of self-sustained discharge results in the effective absorption of microwave field in the region of nonself-sustained discharge, the plasma halo, where a significantly nonequilibrium distribution of temperature characteristics is realized.

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Conflict of interest

The authors declare that they have no conflict of interest.

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