Technical Physics Letters, 2023, Vol. 49, No. 1

04.1

Atmospheric pressure discharge sustained by millimeter radiation in a waveguide plasmatron

© *D.A. Mansfeld, A.V. Vodopyanov, S.V. Sintsov, N.V. Chekmarev, E.I. Preobrazhensky, M.E. Viktorov*

Institute of Applied Physics, Russian Academy of Sciences, Nizhny Novgorod, Russia
E-mail: Mda1981@ipfran.ru

Received October 5, 2022
Revised November 7, 2022
Accepted November 7, 2022

In the developed waveguide plasmatron with combined gas influx, a nonequilibrium discharge maintained at atmospheric pressure by continuous microwave gyrotron radiation at 24 GHz was obtained and studied for the first time. It is shown that in the regime of argon discharge stabilization by carbon dioxide flow the temperature of electrons is 0.2–0.3 eV at gas temperature 1200–1500 K and the electron density does not exceed the cutoff value for the heating frequency in a wide power range.

Keywords: microwave heating, atmospheric pressure discharge, nonequilibrium plasma, gyrotron.

DOI: 10.21883/TPL.2023.01.55345.19384

Non-equilibrium microwave atmospheric-pressure discharge plasma is currently being used more and more often in such technological processes as nondestructive modification and treatment of surfaces of polymer and organic materials by plasma flows and deposition of thin organic films [1]. The processes of decomposition of carbon-dioxide gas (CO_2) [2,3], dry reforming of methane [4], and nitrogen fixation (production of atomic nitrogen) [5] provide no less promising applications for microwave atmospheric-pressure discharge plasma. A wide variety of microwave atmospheric-pressure discharges, which include surface-wave ones (surfatron), discharges with axial gas injection (TIA, TIAGO), and flare-type microwave discharges [6], have been proposed over the last 50 years. Magnetrons with a frequency of 2.45 GHz and a power level ranging from several tens of watts to several kilowatts are used as microwave radiation sources in the overwhelming majority of these discharges. The key issue with the mentioned discharges is that an increase in heating power leads only to an increase in gas temperature and an expansion of the discharge region, while the temperature and density of electrons remain almost unchanged: plasma equilibrates. One of the ways to preserve plasma in non-equilibrium conditions consists in the use of high-power electromagnetic radiation of gyrotrons at frequencies higher than the traditional magnetron frequency of 2.45 GHz. The application of short-wave radiation provides an opportunity to raise the specific energy deposition considerably and thus establish non-equilibrium discharge conditions with the temperature of electrons exceeding the temperature of heavy particles (gas molecules and atoms, excited particles). In addition, higher heating frequencies allow one to increase the plasma density significantly, speeding up plasma-chemical reactions [1].

We have demonstrated and examined in [7] a non-equilibrium (the temperature of electrons exceeded considerably the rotational temperature of gas molecules) discharge mode sustained in a quasi-optical beam of microwave gyrotron radiation with a frequency of 24 GHz. A metallic gas influx tube 6 mm in diameter, which was used to supply plasma-forming gas, was positioned at the microwave beam waist. A microwave discharge (plasma torch up to 5 cm in length with a diameter equal to the tube diameter) was initiated at the gas influx tube exit. A low coefficient of absorption of microwave radiation in plasma (no higher than 10%) is one of the drawbacks of this freely localized discharge. The absorption coefficient was raised to 30–40% in [8] by installing an electrodynamic structure in the shape of a truncated metallic cone in the vicinity of the gas influx tube. This conical structure both enhanced the radiation power density around the beam waist and localized the gas flow in a closed energy release region, thus improving the efficiency of plasma heating. However, the main disadvantages of a conical plasmatron with quasi-optical radiation coupling are relatively high power losses in microwave radiation coupling and focusing units, strict requirements imposed on the adjustment accuracy of the electrodynamic system, and the need to arrange all elements of the electrodynamic system in a tight metal chamber. In the present study, we discuss the design of a waveguide-type microwave plasmatron that has no such drawbacks.

The diagram of the designed waveguide-type plasmatron with combined gas supply is presented in Fig. 1. Microwave heating was performed by continuous-wave electromagnetic radiation of a service gyrotron with a frequency of 24 GHz and a power of 20–5000 W. The plasmatron is a continuation of the superdimensional waveguide of the gyrotron with an inner diameter of 32.6 mm, being connected to it via a vacuum window made of boron nitride. From the design perspective, the waveguide plasmatron features two units: a water cooling unit and a gas supply assembly. In

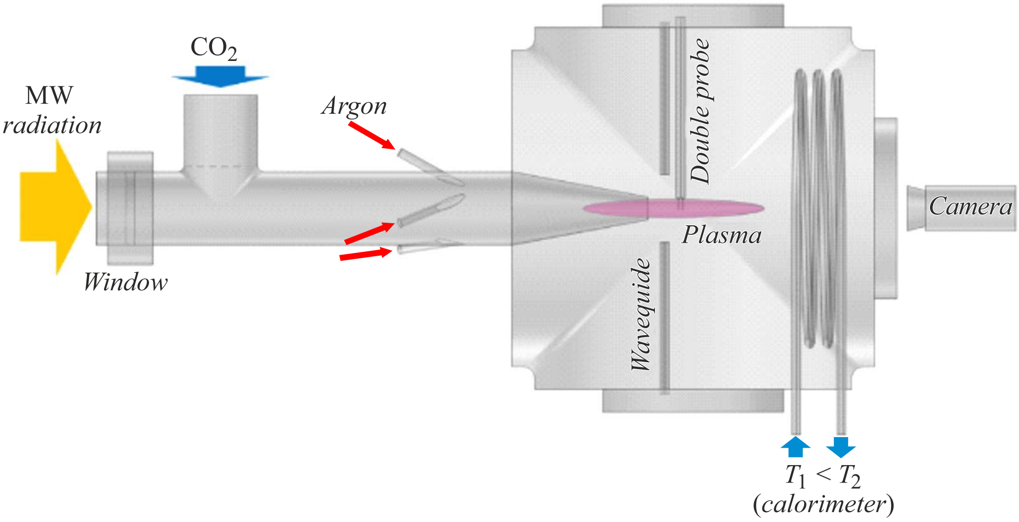

Figure 1. Diagram of the waveguide plasmatron with combined gas supply and a diagnostic expansion chamber.

order to raise the radiation power density, the waveguide is made gradually tapering with a conical horn with a length of 60 mm and an outlet aperture of 10 mm at the end. The plasmatron is connected via a typical CF160 vacuum seal to an expansion chamber in the form of a standard 6-way cross. A flow calorimeter mounted inside the chamber provides an opportunity to measure the power of microwave radiation unabsorbed by the discharge with an accuracy of 0.5 W.

Plasma-forming gas (argon) is supplied via three symmetrically positioned tubes that make an angle of 30° with the plasmatron cylinder element. This tangential gas influx allows one to form swirling gas flows, thus limiting the area of contact of hot plasma with the waveguide walls. The plasmatron is designed so that protective CO_2 gas may also be supplied via a waveguide T-junction from the side of the inlet microwave window. This protective gas supply is required due to the fact that the probability of discharge on the microwave window surface increases with increasing power.

A discharge is initiated by briefly introducing a thin metal wire into the horn aperture at a gyrotron power of approximately 190 W (according to the results of calculations, this corresponds to an electric field intensity of 480 V/cm). A swirling flow of argon under atmospheric pressure is supplied through the gas supply assembly with the use of Bronkhorst flow meters. The optimum argon flow range for discharge initiation is 2–5 l/min. The heating power may be reduced (to a level as low as ~ 20 W) after the initiation of a discharge to protract it. Figures 2, *a* and *b* present photographic images of the optical glow of argon plasma made using a Nanogeit-24 high-sensitivity camera with an exposure time of 100 ns. The camera was aligned with the axis of the system and directed at the horn aperture. At the minimum possible gyrotron power (22 W), the discharge is contained completely within the cone and has the appearance of short-lived filamentary plasma formations that are stretched transversely (Fig. 2, *a*). The coefficient of absorption of microwave radiation by plasma is 30–35%, which is attributable to the small size of filaments. As the power grows through to 190 W, the number of plasma channels increases (Fig. 2, *b*), and they fill the cone volume almost entirely. The plasma torch then extends several millimeters beyond the cone, and the coefficient of absorption of microwave radiation increases through to 80%. When the power grows further, the discharge shifts toward microwave radiation, is initiated near the inlet window, and is extinguished afterwards. Therefore, a stable discharge at power levels in excess of 200–210 W may be observed only if a certain flow of protective gas from the side of the microwave window is added. At the minimum added flow of CO_2 (0.3 l/min), filamentary structures inside the cone become „blurred,“and the discharge assumes a diffuse form (Fig. 2, *c*). The photographic image taken from the side with a common camera (Fig. 2, *d*) reveals that the discharge at the horn outlet is a plasma torch approximately 70 mm in length and 10–15 mm in diameter with a bright hot central part surrounded by excited gas. The geometric dimensions of the plasma torch are defined completely by the ratio of plasma-forming and protective gas flows and the heating power. The absorption coefficient decreases to 70–75%, which may be attributed to a reduction in plasma density on account of additional energy losses due to the excitation of CO_2 molecules. An increase in argon flow has little effect on the discharge pattern, while the addition of CO_2 expands the permissible power range to 1 kW at a CO_2 flow of 2 l/min.

The temperature and density of electrons were measured in the stable discharge mode at an argon flow of 4.25 l/min and a carbon-dioxide gas flow of 0.4 l/min in the plasma torch at a distance of 6 mm from the plasmatron outlet. Temperature measurements were performed by briefly (for no more than 10 s) introducing a double Langmuir probe,

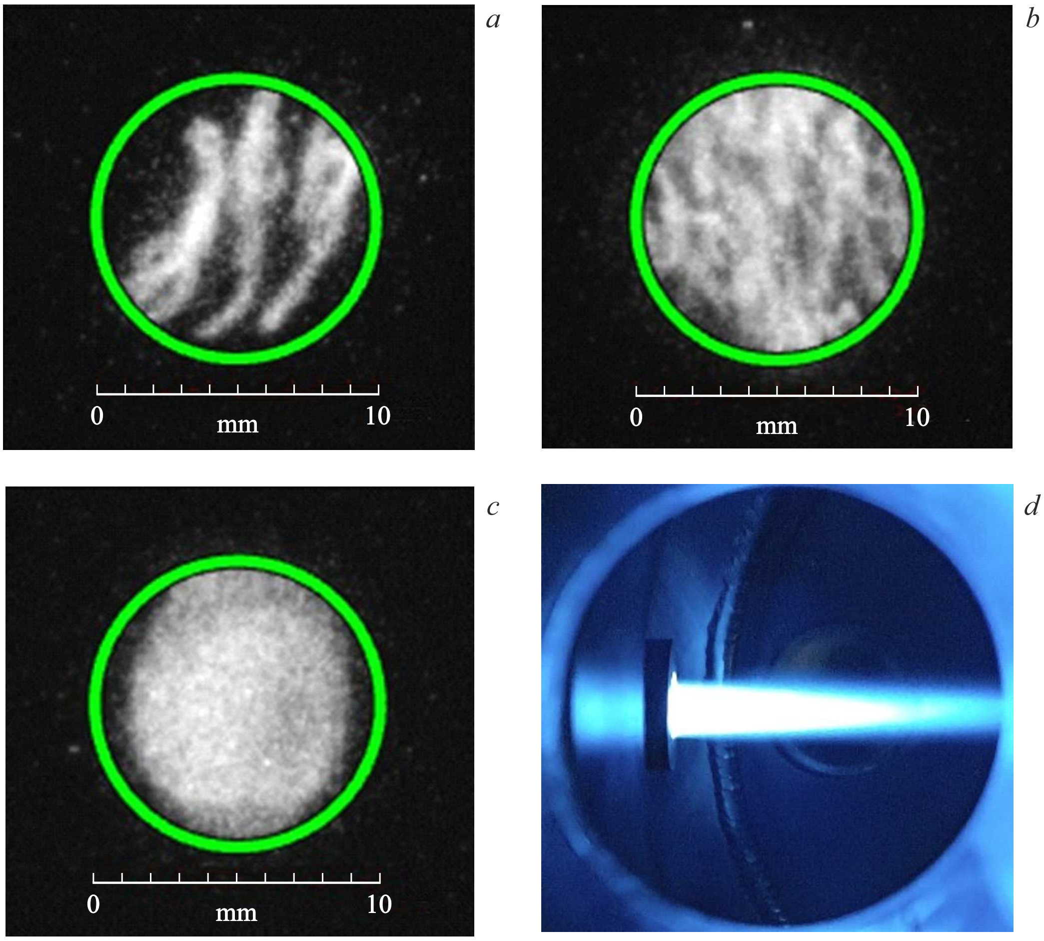

Figure 2. Photographic images of a discharge in argon at a power of ~ 20 (*a*) and 190 W (*b*). *c* and *d* — Discharges in argon and carbon-dioxide gas at a power of 210 W photographed from different perspectives.

which was secured to a movable arm, into the plasma torch. The measured current-voltage curve in the neighborhood of zero is approximated closely by an exponential curve. This provides evidence in favor of the Maxwell–Boltzmann nature of the distribution. It was found that the electron temperature at the copper cone outlet is almost independent of heating power and assumes a value of 0.2–0.3 eV (with an absolute measurement error being equal to 0.05 eV) within the 190–500 W power range. The results of measurements for a non-equilibrium torch were similar [7]. This is apparently attributable to rapid thermalization of plasma at the outlet due to frequent collisions between electrons and neutral particles. The gas temperature was estimated based on the temperature of a fechral wire (measured with an AKIP 9311 pyrometer) introduced into the plasma torch. The obtained gas temperature was 1200–1500 K, which is lower than the electron temperature and is indicative of a non-equilibrium discharge.

The plasma density was determined by direct measurements of the phase incursion of probing radiation with a frequency of 58 GHz passing trough a plasma layer. This method was proven efficient in measurements of the density of chemically active plasma and was discussed in detail in [9]. At a heating power of 200 W, the measured phase incursion was 1.5–2°, which corresponds to an electron density of $(1.9-2.5)\cdot 10^{11}\,\mathrm{cm}^{-3}$ if the characteristic cross-wise plasma size is set to 10 mm. The total error of density measurements did not exceed $0.7\cdot 10^{11}\,\mathrm{cm}^{-3}$. The electron density in the plasma torch increases only slightly at higher power levels and remains well below the critical density value of $7\cdot 10^{12}\,\mathrm{cm}^{-3}$ for a heating frequency of 24 GHz.

Thus, a discharge maintained in the non-equilibrium mode by microwave gyrotron radiation with a frequency of 24 GHz under atmospheric pressure was observed in the plasmatron constructed in the present study. The measured plasma parameters suggest that the plasma torch has potential for application in modern plasma-chemical technology.

Funding

This study was supported by the Russian Science Foundation (project 21-12-00376).

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] M. Narimisa, F. Krčma, Y. Onyshchenko, Z. Kozáková, R. Morent, N. De Geyter, Polymers, **12**, 354 (2020). DOI: 10.3390/polym12020354

[2] Y. Qin, G. Niu, X. Wang, D. Luo, Y. Duan, J. CO_2 Util., **28**, 283 (2018). DOI: 10.1016/j.jcou.2018.10.003

[3] M.Y. Ong, S. Nomanbhay, F. Kusumo, P.L. Show, J. Clean. Prod., **336**, 130447 (2022). DOI: 10.1016/j.jclepro.2022.130447

[4] F. Zhang, X. Zhang, Z. Song, X. Li, X. Zhao, J. Sun, Y. Mao, X. Wang, W. Wang, Fuel, **331**, 125914 (2023). DOI: 10.1016/j.fuel.2022.125914

[5] S. Kelly, A. Bogaerts, Joule, **5**, 3006 (2021). DOI: 10.1016/j.joule.2021.09.009

[6] J. Winter, R. Brandenburg, K.-D. Weltmann, Plasma Sources Sci. Technol., **24**, 064001 (2015). DOI: 10.1088/0963-0252/24/6/064001

[7] S. Sintsov, K. Tabata, D. Mansfeld, A. Vodopyanov, K. Komurasaki, J. Phys. D: Appl. Phys., **53**, 305203 (2020). DOI: 10.1088/1361-6463/ab8999

[8] D. Mansfeld, S. Sintsov, N. Chekmarev, A. Vodopyanov, J. CO_2 Util., **40**, 101197 (2020). DOI: 10.1016/j.jcou.2020.101197

[9] S. Sintsov, D. Mansfeld, E. Preobrazhensky, R. Kornev, N. Chekmarev, M. Viktorov, A. Ermakov, A. Vodopyanov, Plasma Chem. Plasma Process., **42**, 1237 (2022). DOI: 10.1007/s11090-022-10280-0