04

Parameters of the cold plasma jet generated in helium by contact and non-contact initiation

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Comparative studies of the generation of an atmospheric-pressure cold plasma jet in helium excited by a sinusoidal voltage with different methods of its initiation have been carried out. The plasma jet frequency and temperature ranges acceptable for the exposure of biological objects were determined.

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Of a great importance for biomedical applications is non-thermal atmospheric-pressure plasma, for instance, a cold plasma jet (CPJ), that is, a kind of gas discharge able to leave the discharge area under certain conditions. CPJ is a sequence of streamers propagating in the environment in a flow of inert gas being pumped through a discharge The result of the plasma formation impact is device. generation in gaseous and aqueous ion media of nitrogenand oxygen-containing radicals actively interacting with the biological tissue cells. The data of published studies show that active plasma-generated components suppress the vitality of malignant tumors thus selectively affecting civilized and tumor cells (see, e.g. [1-3] and their references). The great variety of conditions and designs of physical-biological experiments, including the diversity of the CPJ generation methods, differences in plasma parameters at the surfaces of targets and various-nature biological objects in vivo and in vitro, as well as the influence of non-controllable factors, complicates the comparison of research results and significantly hinders translation of the developed techniques to model objects and, especially, to living organisms. It seems important to perform comparative investigation of parameters of the plasma jet excited by a sinusoidal voltage in the same discharge device geometry for both the contact and non-contact initiation methods which are most often used in biological experiments. This was the very goal of this study.

The plasma jet was generated by using a source of sinusoidal voltage with controllable frequency $f_U = 10-52$ kHz and amplitude U of up to 7 kV. The voltage magnitude was restricted in order to ensure safe conditions of exposure to CPJ in further experiments with model animals. The streamer breakdown was studied in a flow of 99.995% pure helium. As the plasma jet source, a coaxial dielectric channel 100 mm in length and 10 mm in inner diameter

was used. In the mode of contact initiation (CPJ-CI), the discharge area is confined by the internal potential electrode 50 mm in length and 2 mm in diameter and grounded ring electrode located on the outside of the dielectric channel (Fig. 1, a). In the non-contact initiation mode (CPJ-NI), the discharge area is localized between the metallic cylindrical potential and grounded electrodes mounted outside the channel at the distance of 10 mm (Fig. 1, b); the distance was governed by the maximum permissible voltages. In both cases, at the end of the channel there was mounted a nozzle, that is, a dielectric capillary with the hole diameter of 2.3 mm and length of 5 mm. Voltage U was measured by using an ohmic high-resistance divider. The current was measured with a detector mounted at distance z from the nozzle perpendicular to the CPJ propagation axis; the detector had a form of a collector. Detection of the frequency and amplitude of the current I pulses reaching the collector became possible due to grounding the collector through a low-inductance resistor. As an object of exposure to the plasma jet, we used a target made in the form of an Al₂O₃ ceramic plate 1 mm thick. In all the experiments, the grounded metallic collector was used as an additional electrode for creating in the gap between the plasma streamer and grounded electrode a high-strength electric field configuration. This led to intensification of the active radicals generation in the area where CPJ contacted the target [4]. The distance from the nozzle to substrate was set so as to make the plasma jet touching the target and be typical of biophysical experiments [2,3]. All the experiments were accompanied by measuring the temperature in the region of the plasma jet interaction with the object; for this purpose, thermal imager Testo 872 $\pm 0.1^{\circ}$ C in accuracy was used.



Figure 1. Oscillograms of voltage and current reaching the target. a - CPJ-CI, b - CPJ-NI. 1, 2 - electrodes, 3 - nozzle. <math>v = 9 l/min, $z = 20 \text{ mm}, f_U \approx 52 \text{ kHz}.$



Figure 2. Dependences $I(f_U)$ for modes CPJ-CI (open symbols and crosses) and CPJ-NI (colored symbols). z = 15 mm, v = 9 l/min.

Once the working gas is fed with flow rate v, and sinusoidal voltage with amplitude U and frequency f_U is applied to the electrodes, the plasma jet gets initiated. The CPJ development proceeds in stages well-separated with respect to voltage: (i) discharge formation with subsequent propagation inside the channel with increasing U; (ii) emergence of a plasma jet propagating in free space outside the discharge channel.

In the CPJ-CI mode with the voltage amplitude U = 3-5 kV and flow rate v = 6-9 l/min, the jet length in the absence of the target amounted up to ~ 70 mm. In the CPJ-NI mode with $U \ge 3$ kV and $v \ge 6$ l/min, the jet length did not exceed 30 mm. Figs. 1, *a*, *b* present

typical oscillograms of a single period of voltage and current reaching the collector for the CPJ-CI (a) and CPJ-NI (b) modes.

Fig. 2 presents the frequency characteristics, namely, dependences $I(f_U)$ at v = 91/min and z = 15 mm for the CPJ-CI mode at U = 3.5, 4.3 and 5.2 kV and for the CPJ-NI mode at U = 3, 3.5 and 4.3 kV. The $I(f_U)$ dependences have an intricate shape, which stems from the effect of frequency self-organization of the streamer breakdown. Paper [5] has shown that interaction of the plasma jet generated by the sinusoidal excitation in a helium flow with the target located on the grounded metallic electrode results in that the frequency of touching the target by current f_I

may occur to be lower than the applied voltage frequency f_U by *n* times. The streamers arise and propagate inside the dielectric channel during each voltage cycle; however, while leaving the plasma jet source, the streamer can either reach the target surface or be "extinguished" in interacting with quasi-neutral plasma produced by previous streamers. This effect is governed by the ratio between the density of plasma emerging in the streamer head and residual density of quasi-neutral plasma above the target surface.

The general tendency consists in an increase in amplitude of the current reaching the target with increasing applied-voltage frequency f_U at the same voltages and with increasing U at the same f_U . In the CPJ-CI mode, in the U range of 3-6 kV, current frequency f_I gets proportional to f_U , $f_U/2$ and $f_U/4$ with increasing f_U . Between the regions of a one-to-one correspondence between f_I and f_U there are transient areas with irregular f_I (these areas are hatched). In the CPJ-NI mode at U = 3.5 kV, current frequency f_I is proportional to f_U , $f_U/2$; when $U \ge 4$ kV, a regular character of the current pulses ($f_I = f_U$) is observed; at lower voltages ($U \le 3$ kV), the current frequency behaves as $f_I = f_U/2$.

Fig. 3, *a* presents dependences I(U), namely, the amplitude of current reaching the collector versus voltage, at the gas flow rate v = 91/min. For the CPJ-CI mode, the set of curves is presented for conditions z = 20 mm, $f_U \approx 13 \text{ kHz}$ (these are the conditions under which $f_I = f_U$), $f_U = 26 \text{ kHz}$ ($f_I = f_U/2$) and $f_U = 52 \text{ kHz}$ ($f_I = f_U/4$); for the CPJ-NI mode, the curves for z = 15 mm, $f_U = 15 \text{ kHz}$ ($f_I = f_U$) are given. Dependences I(U) were of the same type in both modes; in the considered gas flow rate range, with increasing U, the current amplitude only slightly depended on v and almost linearly increased with increasing voltage. In the CPJ-CI mode, the achievable current was $I \sim 10-15 \text{ mA}$, that in the CPJ-NI mode did not exceed $I \sim 2-3 \text{ mA}$.

Notice that in the CPJ-CI mode the frequency of current reaching the target remains constant $(f_I = 13 \text{ kHz})$ at voltage frequencies $f_U = 13$, 26 and 52 kHz $(f_I = f_U = 13 \text{ kHz}, f_I = f_U \approx 26/2 \text{ kHz}$ and $f_I = f_U = 52/4 \text{ kHz})$. However, the current amplitude increases with increasing f_U . A similar pattern is observed also in the CPJ-NI mode. Earlier we have shown [6] that the maximal effect of suppressing the cancer cell vitality gets achieved at $f_U = 52 \text{ kHz}$ when the frequency of touching the cells by current remains $f_I \approx 13 \text{ kHz}$. Enhancement of the impact efficiency on the cells is, apparently, ensured by enhancement of currents reaching the biological object.

The therapeutic effect of exposure to plasma depends on the dose and time of the cell treatment, which are restricted by the biological object heating. Evidently, the temperature increase ΔT caused by treatment with CPJ should not result in destabilization of cell proteins. Fig. 3, *b* presents the dependences of the target temperature increment on exposure time $\Delta T(t)$ in the CPJ-CI mode at U = 3.5and 3.9 kV, v = 3 and 91/min, $f_U = 13$ kHz ($f_I \approx f_U$)



Figure 3. a - I(U) dependences. Modes: CPJ-CI (1-3), CPJ-NI (4). v = 91/min; $f_U = 52$ (1), 26 (2), 13 (3) and 15 kHz (4); z = 20 (1-3) and 15 mm (4). b — dependences $\Delta T(t)$. Modes: CPJ-CI (1-6), CPJ-NI (7). v = 9 (1, 2, 5-7), 3 (3) and 61/min (4); $f_U = 13$ (1-3, 7) and 52 kHz (4-6); U = 3.5 (1), 3.9 (2, 3), 2.9 (4, 6), 3.3 (5) and 4 kV (7); z = 20(1-5), 33 (6) and 15 mm (7).

and U = 2.9, and 3.3 kV, v = 6 and 91/min, $f_U = 52 \text{ kHz}$ $(f_I \approx f_U/4)$, and also in the CPJ-NI mode at U = 4 kV, $f_U = 13$ kHz, v = 91/min. The set of experimental results shows that, at the specified parameters of the CPJ initiation, the characteristic time of the target temperature stabilization does not exceed 60 s: in the CPJ-CI mode, the $\Delta T(t)$ dependence is of an almost stationary character, while in the CPJ-NI mode a gradual increase in ΔT is observed. The general tendency is that, when voltage U increases at constant flow rate v and distance z between the nozzle and target (similarly to the case when v decreases at constant U and z or when z decreases at constant U and v), the target temperature increases and can exceed the maximum permissible value for living organisms (taking into account their own temperatures). As the results presented in Fig. 3, a show, at $f_U = 52 \text{ kHz}$ the temperature appears to be higher at a higher achievable current. To reduce ΔT , it is necessary to decrease either the operating voltage or the distance from the nozzle to object, which is not always admissible since leads to a decrease in current with whose magnitude the intensity of active radicals generation is associated. Another way is to increase the working gas flow rate, which in its turn is associated with the deviation from the active radicalsóptimal concentration that is an extremal function of the flow rate [4]. The above is illustrated by dependence $\Delta T(t)$ measured on the skin of mice that were males 8–12 weeks old, line BALB/C, with the mean mass of 25–30 g, grown in the vivarium of Institute for Chemical Biology and Fundamental Medicine, SB RAS (U = 2.9 kV, z = 33 mm, $f_U = 52$ kHz). Under these conditions, $\Delta T \leq 7.5^{\circ}$ C, which is acceptable for working with living objects.

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Compliance with ethical standards

All experiments with animals were performed in accordance with recommendations and requirements for the animal use and welfare (ECC Directive 86/609/EEC). The protocols were approved by the Animal Research Ethics Committee of the SB RAS Administration (Protocol № 61/2 of August 14, 2020).

Conflict of interests

The authors declare that they have no conflict of interests.

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