Phenomena at the electrode surfaces and localization of the volume discharges in small-sized sealed-off TEA-CO$_2$ lasers

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Abstract. The mechanism of localization of volume discharge and limitation of resource in small-sized sealed-off TEA-CO$_2$ lasers are grounded. The main processes which initiate the transition of volume discharge into the local one are connected with autoemissive currents from microinhomogeneities at the cathode surfaces. Maximal resource achieved in TEA-CO$_2$ lasers with electrodes made from materials which more stable to the influence of the volume discharge plasma. There are Mo, Ti, Ta, Nb, W and C (graphite). Lifetime of the small-sized sealed-off TEA-CO$_2$ laser up to 2000 hours using graphite cathodes has been achieved.

1. Introduction

Small-sized sealed-off TEA-CO$_2$ lasers are one of the main components of infrared systems for various purposes [1–4]. One of the main requirements for such TEA-CO$_2$ lasers is to have a significant resource. The creation of small-sized sealed-off TEA-CO$_2$ lasers with a significant resource and stable optical parameters is linked with the solution of two fundamental problems:

- formation of volume discharges resistant to the localization in CO$_2$-laser mixtures containing products of plasma chemical reactions;
- stabilization of energy parameters of laser pulses.

The solution of the first problem have a priority importance, since without the implementation of a spatially homogeneous volume discharge with specified electrical and time parameters, it is impossible to pump and operate the TEA-CO$_2$ lasers themselves. The solution of the second problem is mainly associated with the development of effective methods of regeneration of working mixtures by gas or metal catalysts.

Under the influence of the volume discharge a large number of secondary compounds are formed and accumulated in the working mixture of the sealed TEA-CO$_2$ laser. These compounds initiate the degeneration of the volume pumping discharge into a local one and limit the lifetime of small-sized TEA-CO$_2$ lasers at the level of $10^3$–$10^4$ pulses. Regeneration of working mixtures by means of catalysts allows to increase the lifetime up to $(1–6)\times10^6$ pulses [5]. Localization of the volume discharges after $(1–6)\times10^6$ pulses occurs in conditions of very low degree of dissociation of CO$_2$ and low concentration of molecular oxygen and other secondary active compounds. These facts indicate that the localization of the volume discharge is determined not only by the processes in the gas medium, but, possibly, by the processes on the electrode surface.

This work is devoted to the study of the interrelationships between the microstructure of the working surfaces of the electrodes and the field emission characteristics of these surfaces with the
spatial structure of the volume discharge plasma and the resource of small-sized sealed TEA-CO₂ lasers.

2. Experimental set-up

The structure of the experimental set-up is shown in figure 1. Main elements of the set-up are a gas-discharge chamber (GDC) made from quartz glass, mechanical vacuum pump (MP) and electric discharge high-vacuum (ED) pump, main (PG₁) and auxiliary (PG₂) pulse generators, the synchronizer (S), the system of gas filling of the studied gases (FS), manometer (M) and vacuum gauge (VG). Symbols K₁, K₂, K₃... the vacuum taps are indicated. Additionally, the structure of total set-up includes a high voltage source (HV) with the possibility of smooth adjustment of voltage in the range 0–40 kV and a multi-range microampermeter (µA) with dimensions of measurements in limit 0.01–10 µA. The voltage at the electrode gap at the time of the field measurements of currents were measured by kilovoltmeter (kV).

The gas-discharge chamber could be filled with CO₂-laser mixtures with any ratio between the components of CO₂, N₂ and He up to full pressure up to one atm. It was also possible to fill the discharge chamber by hydrogen, ammonia and nitrogen oxides, which function as gas catalysts.

![Figure 1. Scheme of experimental set-up.](image)

The volume discharge in four interelectrode gaps was ignited from high-voltage pulse generators in CO₂-laser mixtures. For each of the discharge intervals an individual capacitive energy storage capacitor was switched. Regulation of the storage energy inject into the volume discharge plasma can regulated by changing the capacity of the individual storage capacitor or changing the voltage of its charge. Preliminary ionization of gas mixtures in all four intervals was simultaneously carried out by UV radiation of an auxiliary surface discharge, which was excited by an autonomous pulse generator (PG₂). The starting moments of the auxiliary and volume discharges were synchronized with an external generator (S).

The study of the spatial structure of the volume discharge and the autoelectronic characteristics of the cathodes was made in a gas-discharge chamber made of quartz glass (figure 2), in which there were identical in shape cathode (C₁–C₄) and one flat anode (A) of nickel. The interelectrode gap in each pair of electrodes could simultaneously changed by vacuum mechanical unit (SU) in the range from fractions of a millimeter for two centimeters. For the volume discharge ignition the interelectrode gap values were set equal to 1 cm. At the autoelectronic measurements the magnitude of the interelectrode gaps was decreased up to 1 mm. The distance between the cathodes was set to 2 cm. This distance was enough to exclude mutual influence effects of discharge gaps on each other. The cathodes were made of different materials and had the shape of a Bruce profile [6]. Al, Ni, Cu, Mo, Ta, Nb, W and C (graphite) were used as electrode materials.

Volume discharges between electrodes made of different materials were ignited in CO₂-laser mixtures in the intervals with the interelectrode distance d = 1 cm. An individual storage capacitor was
switched for each discharge gap. Volume discharges of nanosecond duration with energy density \( W = 150–250 \text{ mJ/cm}^3 \) were simultaneously excited in all intervals at pulse repetition frequency \( F = 25 \text{ Hz} \). This pulse repetition rate was provided by the use of "electrical wind" effect [7] inside the gas-discharge chamber (electrode system for the formation of "electrical wind" in figure 2 not shown.

![Figure 2. The structure of the gas discharge chamber.](image)

Before studies of the volume discharge formation in CO\(_2\)-laser mixtures for a long time, the gas-discharge chamber was pumped out by the mechanical vacuum pump (MP) and electric discharge (ED) pump to a residual pressure \( P < 10^{-7} \text{ Torr} \). The pressure of the residual gases was controlled by the vacuum gauge VIT-2 (VG). After that, the interelectrode interval was set to 1 mm by means of a vacuum mechanical unit (SU) and the dependences of the autoelectronic currents on the applied voltage to the electrodes were measured. The same procedure was performed after a certain number of discharge pulses and after the completion of studies of the volume discharge spatial structure.

The obtained dependences of the autoelectronic currents on the voltage were then translated by the Fowler-Nordheim method [8, 9] into "modified" coordinates, which, according to the known value of the electron work function from the surface of the material under study, make it possible to determine the value of the local amplification coefficient of the electric field. The values of the electron work function from the working surfaces of the electrodes were taken from the reference literature [10–12] and independently measured by the method of "vibrating capacitor" in a separate high–vacuum chamber.

Carrying out of autoelectronic measurements before the ignition of the volume discharge, after some time after switch-on of volume discharges (after a certain number of discharge pulses) and after the completion of the whole process of monitoring the spatial structure of the volume discharge allows to control the dynamics of the local electric field gain on the micro-homogeneity of the cathode surface from various materials changing under the influence of the space discharge plasma. The data obtained in this way of the local electric field gain behavior can be useful both in the analysis of the process of degeneration of the volume discharge into a local one in these circumstances, and in the foundation of new mechanisms of localization of the volume discharge in the active media of small-sized sealed-off TEA-CO\(_2\) lasers. In addition, information on the behavior of the local amplification coefficient of the electric field can be used as a basis for the analytical prediction of the volume discharge resource and the resource of small-sized sealed-off TEA-CO\(_2\) lasers.

After the autoelectronic measurements the interelectrode gap was set to 1 cm, the gas-discharge chamber was filled with a CO\(_2\)-laser mixture and volume discharges were formed in the all discharge gaps. The spatial structure of the space discharges was controlled visually and photographically in each of the intervals. If in some gaps the volume discharge was transformed in the local that gap was disconnected and the excitation of the volume discharges in other gaps has been continuing.
The state of the working surfaces of the electrodes before influence on them of the volume discharge plasma and after action a certain number of discharges pulses the surface structure was controlled by a scanning electron microscope JEOL JSM-6610.

3. Research results
The volume discharge current (pumping current) in the CO\textsubscript{2}-laser mixture (CO\textsubscript{2}:N\textsubscript{2}:He) leads to a noticeable change in the initial chemical composition and the appearance of a big number secondary compounds [5, 13]. The main component of these secondary compounds and has the most chemical activity is oxygen. Its concentration in the equilibrium state can reach several percent.

Prolonged influence of the volume discharge plasma containing oxygen and oxygen compounds in excited and ionized states on the surface of the electrodes leads to the formation of oxide films on them and a change in the microstructure of the surfaces.

Changes in the microstructure of the surfaces of Al, Ni, Ta, and C (graphite) electrodes under the influence of the volume discharge plasma in a CO\textsubscript{2}:N\textsubscript{2}:He = 1:1:8 mixture are shown in figure 3. The scale of the images – M = 500:1. The absolute values of the geometric dimensions in the photographs are 160 µm×160 µm. The top row corresponds to “clean” surfaces, i.e. surfaces that have not been interaction with volume discharge plasma. The bottom row reflects the state of the surfaces of the electrodes, which were exposed at 10\textsuperscript{6} discharge pulses. It is seen that the effect of a plasma of a volume discharge leads to noticeable changes in the microstructure of the surfaces of the cathodes. They are covered with dense layers, apparently, of oxide compounds. The thickness of these oxide layers measured by micrometer reaches values up to Δ ~ 0.1–0.2 mm.

![Figure 3](image_url)

**Figure 3.** The microstructure of the surface of the electrodes from Al, Ni, Ta and graphite (C) before exposure to plasma space discharge (top row) and after 10\textsuperscript{6} discharge pulses (bottom row).

Visual observations of the spatial structure of the volume discharges in the gas-discharge gaps showed that the working surface of the cathode is completely covered with brightly glowing filamentous plasma formations. The distance between them on the cathode surface is ≈ 1 mm and it changes very little over time. The diameter of the filamentous plasma formations (in the cathode region of the discharge) is visually estimated to be a fraction of a millimeter. The length of these luminous formations increases with time in the direction of the anode. When the length of the filamentous formations reaches a value of 30–40% from the interelectrode interval, one of the filamentous channels begins to overlap the entire interval and then only a high–current luminous local discharge is formed in the interelectrode interval. Such discharges are not suitable for excitation of laser mixtures and creative an active medium.

In the figure 4 fragments of the volume discharge glow in the gaps formed by profiled electrodes from aluminum, nickel, tantalum and graphite (C) with the same shape and geometric dimensions are presented. The upper row of photos shows the glow of the volume discharge in freshly prepared CO\textsubscript{2}-
laser mixtures, the lower row- the glow of the volume discharge in the same gas-discharge intervals after $10^6$ discharge pulses. Cathodes on the all photos are located below.

The structure of the plasma of the volume discharge in all intervals in the initial mixture of gases has a high spatial homogeneity with an increased luminescence brightness in the cathode region. The filamentous channels formed in the cathode region increase in length towards to the anode as the number of discharge pulses increases.

In the discharge gaps with cathodes from aluminum after $10^6$ discharge pulses observed bright-luminous high-current channels covering the entire discharge gap. The pumping laser in the plasma of such local discharge is impossible. In gas-discharge gaps with cathodes from Ni, Ta and C (graphite) the discharge structure does not change during the same time. The smallest changes in the structure of the volume discharge in the cathode region and in the positive column are observed in the gaps from Ni and Ta.

**Figure 4.** The spatial structure of the volume discharge in the gaps with cathodes from Al, Ni, Ta and C (graphite) in freshly prepared CO$_2$-laser mixtures (upper row) and in equilibrium mixtures (lower row) after $10^6$ discharge pulses.

Thereby the method of experimental determination of the volume discharge resource consisted in visual observation of the volume discharge plasma and in the registration of the time interval within which the filamentous channels will begin to overlap the interelectrode interval and this will be followed by a complete degeneration of the spatially homogeneous volume discharge into a high-current local discharge not suitable for pumping purposes.

Changes in the microstructure of the electrode active surfaces result in a multiple increase of up to 10 times or more in the autoelectronic currents. The volt-ampere characteristics of autoelectronic currents, formed in Fowler-Nordheim coordinates, easily allow to determine the electrical field amplification coefficient $\beta$. These characteristics are shown in the figure 5. The initial values of electrical field amplification coefficient $\beta_0$ for Al, Ni, Mo, W, Ta and graphite have the next magnitudes – 570, 530, 700, 340, 740 and 420, correspondingly. The final values of that coefficient $\beta_t$ are 1650, 1240, 704, 240, 500 and 410.
Figure 5. Fowler-Nordheim volt-ampere characteristics of the autoelectronic currents from cathodes made from Al, Ni, Mo, W, Ta and graphite before influence of the volume discharge plasma on them (1) and after $10^6$ discharge pulses (2).

The values of that coefficient are increased in the same materials, where the most noticeable changes on the surface microstructure are observed. These materials are aluminum, copper and nickel. The value of the amplification coefficient on the electrodes made of molybdenum, tantalum, tungsten and graphite are changed insignificantly.

Increase in geometrical dimensions of microinhomogeneities at the electrode surfaces results in increase autoelectronic currents from these zones of the surface and initiates a formation of emission centers on them as a result of a transfer from autoelectronic emission to an explosive one [14, 15].

The measurements of the electron emissivity energy prior to the effect of a volume discharge plasma on the electrode surfaces and after $10^6$ discharge pulses showed that the electron emissivity energy changes only to 0.05–0.2 eV.

The comparison of autoelectronic current values coming out of the investigated material surfaces while changing the electron emissivity energy to 0.05–0.2 eV together with corresponding current changes, in the case of increasing the amplification coefficient, indicates that the increase of autoelectronic currents are mostly connected with the increase of the local values of electrical fields occurring on the micro–heterogeneities being formed.

The emission centers formed on the electrode surfaces initiate strongly conducting plasma formations in the form of visually luminous filaments. The length of these threads increases over time (with an increase in the number of generated discharge pulses). Figure 6 demonstrates dependencies of the thread-like channels length from the total time of the volume discharge interaction with the electrode surfaces at the pulse repetition rate $F = 25$ Hz. The vertical axis shows the length of the filamentous channels in millimeters. The value of $Z = 8$ mm corresponds to the value of the interelectrode gap. The length of the filamentous channels was determined by the enlarged optical image on the screen with a millimeter grid.
Figure 6. Dependences of the length of filamentary channels from the burning time of a volume discharge at a pulse repetition rate of $F_{RP} = 25$ Hz for various cathode materials. Working gas mixture $CO_2:N_2:He = 1:1:6$, total pressure of the mixture $P_{\Sigma} = 1$ atm.

Aluminum, nickel, tantalum and graphite (C) were used as the cathode materials. The volume discharge in all cases was ignited at a pulse repetition rate of 25 Hz. The initial length of the filamentous channels in the cathode region is ~ 1 mm. As the number of discharge pulses increases the length of the filamentous channels increases. This increase is insignificant in some areas of the above dependencies and is rapidly increasing in others.

The total operating time of the gas-discharge device after which there is a rapid increase in the length of the filamentous channels and the subsequent complete overlap of the gap and the formation of only one high-current local arc channel in it is determined (under other identical conditions) by the cathode material.

After ~ $10^6$ discharge pulses, the filamentous channels completely cover the discharge gap with an aluminum cathode. In the intervals with Ni and Ta cathodes after $10^6$ discharge pulses the linear dimensions of the filamentous channels reach ~ 20–40% of the interelectrode gap. In the gaps with the cathodes of molybdenum, tantalum, niobium, tungsten and graphite the spatial structure of the volume discharge plasma after $10^6$ pulses does not undergo any visible changes. The filamentous channels formed in the cathode region of the volume discharge retain their geometric position relative to the cathode surface as the number of discharge pulses increases.

The effect of the volume discharge plasma on the working surfaces of the electrodes leads to a decrease in the electron work function from these surfaces on the 0.1–0.2 eV. The observed changes in the electron work function from the working surfaces may be associated with the formation of oxide compounds, salts, carbides, nitrides, mixed oxides, alloys with carbon monoxide and a number of other compounds [10, 11]. The values of the electron work function of these compounds vary within a wider range than that of pure metals [12].

The greatest changes in the gain of the electric field on the micro-inhomogeneities of the cathode surface after exposure to the plasma of the volume discharge are observed in aluminum, nickel and copper. The magnitude of the electric field amplification on the surfaces of electrodes made of molybdenum, tantalum, niobium, tungsten and graphite varies slightly.

The obtained results show that the increase in the autoelectronic currents from the cathode surface is mainly due to the increase in the local values of the electric field intensity on the micro inhomogeneities formed under the influence of the volume discharge plasma. These results also indicate on the manufacturing of electrodes of small-sized sealed TEA-CO$_2$ lasers from materials such as Mo, W, Ta, Nb and graphite.
Overfilling of the discharge chamber after $10^6$ discharge pulses with a fresh working mixture and repeated studies of the spatial structure of the volume discharge with electrodes whose working surface has already been subjected to prolonged exposure to gas-discharge plasma showed that local channels in the cathode region of the volume discharge appear (depending on the type of electrode material) after $10^3$–$10^4$ pulses in the same places of the cathodes where they were recorded in the previous series of measurements.

Shown in figure 6 dependences of the length of the filamentous channels from the duration of plasma exposure to the surface of the electrodes show that the volume discharge in the gaps with aluminum electrodes retains its spatial homogeneity during 8–10 hours. For the discharge gaps with a nickel cathode, the life of the volume discharge reaches 50–80 hours. Resource $T = 280$–300 hours is achieved in case of the manufacturing of cathodes from Ta and Nb. The greatest resource (up to 2000 hours) was achieved in the small-sized sealed TEA-CO$_2$ laser with graphite cathode.

4. Conclusion
The obtained results show that the time during which the volume discharge retains its spatial homogeneity (the lifetime of the volume discharge) in CO$_2$-laser mixtures at atmospheric pressure is determined by the kind of cathode material.

As the interaction time of the volume discharge plasma with these surfaces increases, changes in the structure of the working surfaces of the cathodes, an increase in the autoelectronic currents from these surfaces, and an increase in the length of the filamentous channels are observed. The filamentous channels begin their growth from certain points of the cathode surface and do not migrate along the surface over time. These facts indicate that emission centers are formed on the surface of the cathodes and their emission capacity increases as the plasma of the volume discharge affects the working surfaces of the cathodes. When a certain critical emission level is reached, the current of the local channel becomes such that a streamer with a strongly conducting channel is formed over the period of electron drift through the interelectrode gap.

The time during which such changes occur is determined by the nature of the cathode material. The most suitable materials for electrode manufacturing small-sized sealed-off TEA-CO$_2$ lasers are tantalum, niobium, tungsten and graphite. A special place among the materials for the manufacture of cathodes belongs to graphite. Using graphite electrodes the lifetime of a small-sized sealed TEA-CO$_2$ laser at the level of $T \geq 2000$ hours was achieved.

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